PB84-163286

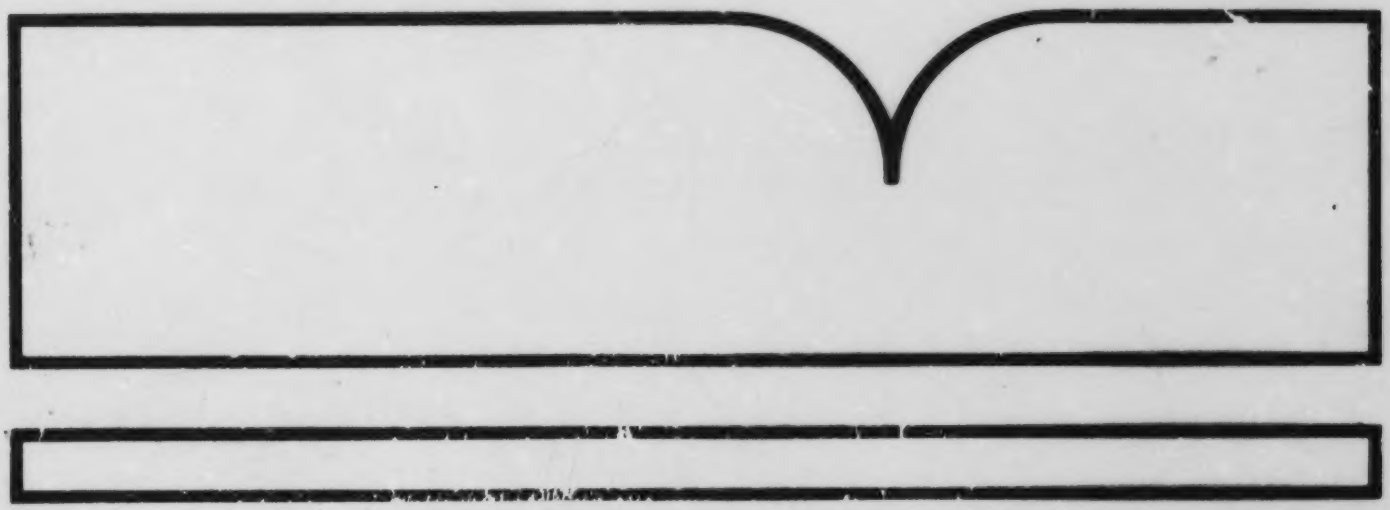
Pollutant Formation during Fixed-Bed and Suspension Coal Combustion

Utah Univ., Salt Lake City

Prepared for

Industrial Environmental Research Lab.
Research Triangle Park, NC

Feb 84



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National Technical Information Service
NTIS

PB84-163286

EPA-600/7-84-026
February 1984

POLLUTANT FORMATION DURING FIXED-BED AND SUSPENSION COAL COMBUSTION

by

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TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO.	2.	3. RECIPIENT'S ACCESSION NO.
EPA-600/7-84-026		PB84 163286

4. TITLE AND SUBTITLE	5. REPORT DATE
Pollutant Formation During Fixed-bed and Suspension Coal Combustion	February 1984
	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) S. C. Manis, J. M. Munro, S. P. Purcell, G. P. Starley, D. M. Slaughter, and D. W. Pershing	8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS Department of Chemical Engineering University of Utah Salt Lake City, Utah 84112	10. PROGRAM ELEMENT NO.
	11. CONTRACT/GRANT NO. R805899
12. SPONSORING AGENCY NAME AND ADDRESS EPA, Office of Research and Development Industrial Environmental Research Laboratory Research Triangle Park, NC 27711	13. TYPE OF REPORT AND PERIOD COVERED Final; 6/78-6/81
	14. SPONSORING AGENCY CODE EPA/600/13

15. SUPPLEMENTARY NOTES IERL-RTP project officer is G. Blair Martin, Mail Drop 62-B, 919/541-7504.

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17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Pollution	Pollution Control	13B
Coal	Stationary Sources	21D
Combustion		21B
Nitrogen Oxides		07B
Sulfur Oxides		
Stokers		13A

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES
Release to Public	Unclassified	79
	20. SECURITY CLASS (This page) Unclassified	22. PRICE

EPA Form 2220-1 (9-73)

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ABSTRACT

This report summarizes results of a 3-year laboratory study of the formation and control of nitrogen and sulfur oxides (NO_x and SO_x) in industrial coal-fired boilers, with emphasis on stoker-coal units. Three major research areas were considered: the evolution and oxidation of fuel nitrogen and sulfur, the retention of SO_x by ash and/or solid-chemical sorbents, and the effectiveness of distributed air addition for NO_x control. The study also included quantification of the combustion process in a stoker environment and consideration of possible detrimental effects of control technology on boiler operation.

Three laboratory furnaces were used to investigate the various combustion regimes of major importance in stoker-fired systems: (1) a 200,000 Btu/hr tower furnace was used to characterize the suspension-phase oxidation of fuel nitrogen and sulfur as a function of local oxygen concentration, temperature, and particle size; (2) a 300,000 Btu/hr fixed-bed furnace was used to study NO_x and SO_x formation in mass-burning units as a function of stoichiometry distribution, burning rate, and overfire-air height (coal/limestone pellets for SO_x control were also considered in this system); and (3) a 750,000 Btu/hr model spreader-stoker furnace was used to investigate NO_x formation and control in a system where both suspension-phase and bed combustion are significant. All of the experimental studies were conducted with high volatile bituminous coals from Utah (low sulfur) and Indiana (high sulfur).

Study results indicate that two types of NO_x controls appear to be viable: (1) the coal feed should be screened to remove the fines (particles less than 0.1 in.), essentially eliminating the high conversion of nitrogen evolved in the suspension zone and resulting in an overall emissions reduction of about 10-40%, depending on the amount of fines normally present in the raw coal; or (2) the primary overfire-air injection port could be moved to above the spreader, and the suspension zone and bed region could both be operated substoichiometric, reducing emissions by as much as 50%. Unfortunately, controlling SO_2 formation in either a spreader or mass-burning stoker unit appears to be difficult. Coal/limestone pellets can significantly reduce SO_2 emissions, and the effectiveness of the sorbent appears to increase when the bed-region stoichiometry is reduced below 1.0.

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TABLE OF CONTENTS

	PAGE
ABSTRACT	iii
LIST OF FIGURES	vi
LIST OF TABLES	viii
CHAPTER	
1. INTRODUCTION	1
BACKGROUND	1
PURPOSE AND SCOPE	4
2. EXPERIMENTAL SYSTEMS	6
TOWER FURNACE	6
FIXED-BED FURNACE	9
MODEL SPREADER-STOKER FURNACE	12
FUEL ANALYSIS	14
3. SUSPENSION-PHASE COMBUSTION	18
FATE OF MAJOR CONSTITUENTS	21
Carbon	21
Hydrogen	25
Nitrogen	25
INFLUENCE OF PROCESS VARIABLES	27
Carbon	28
Hydrogen	28
Nitrogen	30
Sulfur	33

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BEGIN LAST LINE OF TEXT
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PAGE NUMBER
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EPA Form 2350-4 (4-80)
(PREVIOUSLY CIN, EPA FORM 287)
TYPING GUIDE SHEET

Table of Contents (Continued)

CONCLUSIONS. 38

Hydrogen Oxidation. 38

Nitrogen Oxidation. 38

Sulfur Oxidation. 38

4. FIXED-BED STUDIES 39

TYPICAL EXPERIMENTAL RESULTS 39

Excess Air Combustion 39

Staged Combustion 41

INFLUENCE OF COMBUSTION PARAMETERS 44

Air Distribution--Bed Region Stoichiometry. 45

Air Distribution--Excess Air Addition 45

Overfire-Air Height 48

Burning Rate 50

COAL-LIMESTONE PELLETS 53

CONCLUSIONS 57

NO Formation and Control. 57

SO_2 Formation and Control 59

5. MODEL SPREADER-STOKER STUDIES 60

COMPARISON WITH FIELD DATA 60

TOTAL STOICHIOMETRY. 62

BED-REGION STOICHIOMETRY 62

OVERFIRE AIR HEIGHT. 65

6. DISCUSSION. 67

REFERENCES 69

EPA Form 2350-4 (4-80)
(PREVIOUSLY CIN. EPA FORM 287)

TYPING GUIDE SHEET

LIST OF FIGURES

Number		Page
2- 1	Tower furnace Schematic.	7
2- 2	Sideview of the fixed-bed furnace.	10
2- 3	Model stoker furnace cross-section	13
2- 4	Influence of raw coal particle size on distribution of sulfur forms	16
3- 1	Stoker coal size distribution.	19
3- 2	Large particle results	20
3- 3	Fate of carbon and hydrogen in the tower furnace for particles <.063" with 1900°F thermal environment and 670,000 Btu/hr/ft^2 SBHRR	22
3- 4	Fate of nitrogen and sulfur in the tower furnace for particles <.063" with 1900°F thermal environment and 670,000 Btu/hr/ft^2 SBHRR	23
3- 5	Distribution of sulfur forms in exhaust and bottom solids for coal particles <.063" and 1900°F thermal environment.	24
3- 6	Influence of combustion parameters - thermal environment, oxygen partial pressure and coal particle size on carbon and hydrogen evolution	29
3- 7	Influence of combustion parameters-thermal environment, oxygen partial pressure, and particle size on evolution of nitrogen.	31
3- 8	Solid nitrogen analysis for material obtained in bottom and exhaust solid collectors after combustion of particles.	32
3- 9	Influence of combustion parameters - thermal environment, oxygen partial pressure and particle size on oxidation of total sulfur.	34

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TYPING GUIDE SHEET

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List of Figures (Continued)

3-10	Influence of oxygen partial pressure on sulfur and carbon oxidation.	36
3-11	Solid sulfur analysis for material obtained in bottom and exhaust solid collectors after combustion of particles <.063 in	37
4- 1	Burning rate and NO emissions for a Utah coal at SR_b = 1.35.	40
4- 2	SO_2 emissions for a Utah coal at SR_b - SR_t = 1.35. . .	42
4-3a	Overall and bed-region stoichiometries	43
4-3b	Superficial burning rate and exhaust NO emissions. . .	43
4- 4	Effect of bed-region stoichiometry on NO_x emissions. .	46
4- 5	Effect of excess oxygen on NO_x emissions	47
4- 6	Effect of overfire-air height on NO_x emissions	49
4- 7	Effect of superficial burning rate on NO_x emissions. .	51
4- 8	Normalized SO_2/CO_2 evolution for a Utah coal at SR_b = .7	52
4- 9	Post maximum burning rate correlation for various bed stoichiometries	54
4-10	SO_2 emissions and burning rate for Battelle pellets. .	56
4-11	Integrated SO_2 emissions for batch mode burning of Battelle pellets	58
5- 1	Field data comparison	61
5- 2	Effect of overfire air.	63
5- 3	Effect of bed stoichiometry	64
5- 4	Effect of overfire air height	66

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LIST OF TABLES

Number		Page
2-1	Fuel Analysis.	15
4-1	Ultimate Analysis of Battelle Pellets.	55

BEGIN FIRST LINE OF TEXT HERE
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CHAPTER 1

INTRODUCTION

BACKGROUND

In recent years, this country has experienced a renewed emphasis on the use of coal as an energy alternative to imported oil. Considerable research is being directed at various conversion schemes in which coal may be converted to liquid or gaseous fuels. While it is imperative that these processes be developed, it is also a virtual certainty that, for the next one or two decades, the primary mode of coal utilization will be direct combustion. Considerable research has been directed to combustion of pulverized coal and its associated environmental problems. However, many current industrial and commercial boilers are stoker-fired systems; these systems have been largely overlooked in energy and environmental research programs.

At present, stoker-fired boilers are significant both in terms of coal consumption and environmental impact. In 1974 almost 20 percent of the coal consumed in the United States was burned in stoker systems. Stoker fired, water-tube boilers are the fourth largest source of SO_x emissions and account for 13 percent of the NO_x emissions from all coal-fired systems.[1] The related environmental impact may be even more significant than indicated by mass emissions because stoker fired furnaces are often located in congested metropolitan areas. Further, the energy and environmental importance of stoker coal-firing seems certain to grow with the increase in industrial coal utilization that is projected. It is important, therefore, that further

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knowledge be obtained concerning the formation and control of nitrogen and sulfur oxides under conditions typical of stoker firing.

Stoker-fired boilers differ from pulverized-coal-fired systems in physical configuration, mechanisms of particle ignition and burnout, and commercial utilization. In pulverized-coal-fired boilers, the fuel is fed as very fine particles (approximately 70 percent less than 200-mesh) and combustion occurs entirely in the suspension phase. In contrast, stoker systems utilize a range of fuel particle sizes (from fines through large 1-1/2 inch pebbles) with a mean of approximately 3/4 inch, and at least half of the combustion occurs in a solid fuel bed lying beneath the suspension phase. The differing combustion mechanisms occurring in stoker furnaces are a direct consequence of the much larger fuel particles and the combustion regimes in which the particles devolatize and burn (the overthrow phase and the solid fuel bed).

Four main groups of stokers are common in the United States:

- spreader-stokers (overthrow feed);
- underfeed stokers (single and multiple retort);
- water-cooled vibrating-grate stokers (overfeed from hopper); and
- chain-grate and traveling-grate stokers (overfeed from hoppers).

Spreader stokers are characterized by a combination of suspension and fixed-bed burning and are usually specified in boilers with capacities ranging from 40,000 to 400,000 pph steam. The other three types of stokers are collectively referred to as mass-burning stokers and are characterized by thick, solid-fuel beds and the absence of an overthrow phase. In general, this class of stokers is specified in boilers of the range of 15,000 to 250,000 pph steam.

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Recent large-scale pilot and field tests have partially demonstrated the potential of combustion modifications for NO_x control. Giammar and co-workers[2] have conducted tests on a 25,000 pph steam spreader-stoker with a water-cooled vibrating grate and found that decreased NO_x emissions could be obtained by decreasing the overall excess air addition (at a constant load). Gabrielson and Langsjoen[3] tested several large-scale industrial stokers and reported little effect of overfire-air percentage on NO_x emissions and an apparent increase in NO_x emissions with increasing load. Laboratory experiments in a 180mm pot furnace (batch operation) have suggested that the oxidation of volatile fuel nitrogen may be of major importance in stoker systems, particularly in the early portion of the burning.[4] However, the fundamental mechanisms of NO_x formation and parameters controlling NO_x emissions are not well defined.

The information on sulfur oxidation and control in these systems is very limited. Coal sulfur is generally divided into three groups:[5] sulfate sulfur, pyritic sulfur, and organic sulfur. The available inert pyrolysis data show that the organic sulfur is more volatile than coal carbon or nitrogen but less volatile than oxygen or hydrogen, [6, 7] and that the inorganic sulfur can interact with the organic matrix.[5, 6]

Limestone addition appears to be the most attractive method of SO_2 control in a stoker-fired system. This process involves the calcination of $CaCO_3$ and subsequent reaction to form calcium sulfate.[8] Fundamental and pilot-scaled data[9-12] have indicated the viability of the concept and demonstrated the importance of particle size and thermal environment. Recent results[13, 14] have emphasized the importance of the carbon chemistry and the local stoichiometry.

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PURPOSE AND SCOPE

This report summarizes the results of a three-year research program funded by the U.S. Environmental Protection Agency under Grant R-805899 to study the formation and control of nitrogen and sulfur oxides in industrial, coal-fired boilers with emphasis on stoker-fired units. In particular, this program considered the following major research areas:

1. the evolution and oxidation of fuel nitrogen and sulfur;
2. the retention of sulfur oxides by ash and/or solid-chemical sorbents;
3. the effectivness of distributed air addition for NO_x control.

In addition, the study attempted to quantify the combustion process in a stoker environment and considered possible detrimental effects of control technology on boiler operation. The work considered conditions typical of both spreader-stokers and mass-burning systems.

The approach was primarily experimental; three types of laboratory furnaces were utilized to investigate the various combustion regimes of major importance in stoker-fired systems:

1. A 200,000 Btu/hr tower furnace was used to characterize the suspension phase oxidation of fuel nitrogen and sulfur as a function of local oxygen concentration, temperature, and particle size.
2. A 300,000 Btu/hr fixed-bed furnace was used to study NO_x and SO_x formation in mass-burning units as a function of stoichiometry distribution, burning rate, and overfire-air

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height. The effectiveness of coal-limestone pellets for SO_x control was also considered in this system.

3. A 750,000 Btu/hr model spreader-stoker furnace was used to investigate NO_x formation and control in a system where both suspension-phase and bed combustion are significant.

All of the experimental studies were conducted with high volatile bituminous coals from Utah (low sulfur) and Indiana (high sulfur).

The next section of this report describes each of the small-scale furnaces, the associated experimental procedures, and fuel characteristics. The following three sections then summarizes the findings from each of the experimental studies. The final section of the report considers the overall practical implications of the work.

Complete details of all of the experimental systems (including fabrication drawings), analytical methods, and experimental data are available.

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CHAPTER 2

EXPERIMENTAL SYSTEMS

TOWER FURNACE

The experimental furnace used in the suspension combustion studies is illustrated in Fig. 2-1. The furnace was designed to simulate the environment experienced by a particle during the suspension-burning phase of combustion in a spreader-stoker. The vertical combustion chamber is 9 feet high with an inside diameter of 6 inches. The outside dimension of the combustion facility is approximately 28 inches square with a height of 12 feet. The furnace consists of an outer steel shell, 4 inches of high-temperature board insulation, 5 inches of lightweight castable refractory and 2 inches of high-temperature castable refractory. There are numerous 2- and 3-inch observations ports located over the length of the furnace to allow flame and particle photography, visual observation, and optical wall and combustion gas temperature measurements.

The main natural-gas-fired burner is attached to the horizontal extension at the bottom of the furnace. The main burner could provide a full-load firing rate of 215,000 Btu/hr; the hot combustion products produced flowed up vertically to simulate the combustion products leaving the actual stoker bed.

To provide independent wall-temperature control, a pair of 100,000 Btu/hr, natural-gas auxiliary burners were attached to the upper portion of the furnace. The auxiliary burners maintained a constant wall temperature by firing natural gas into the annular channels around the main combustion chamber.

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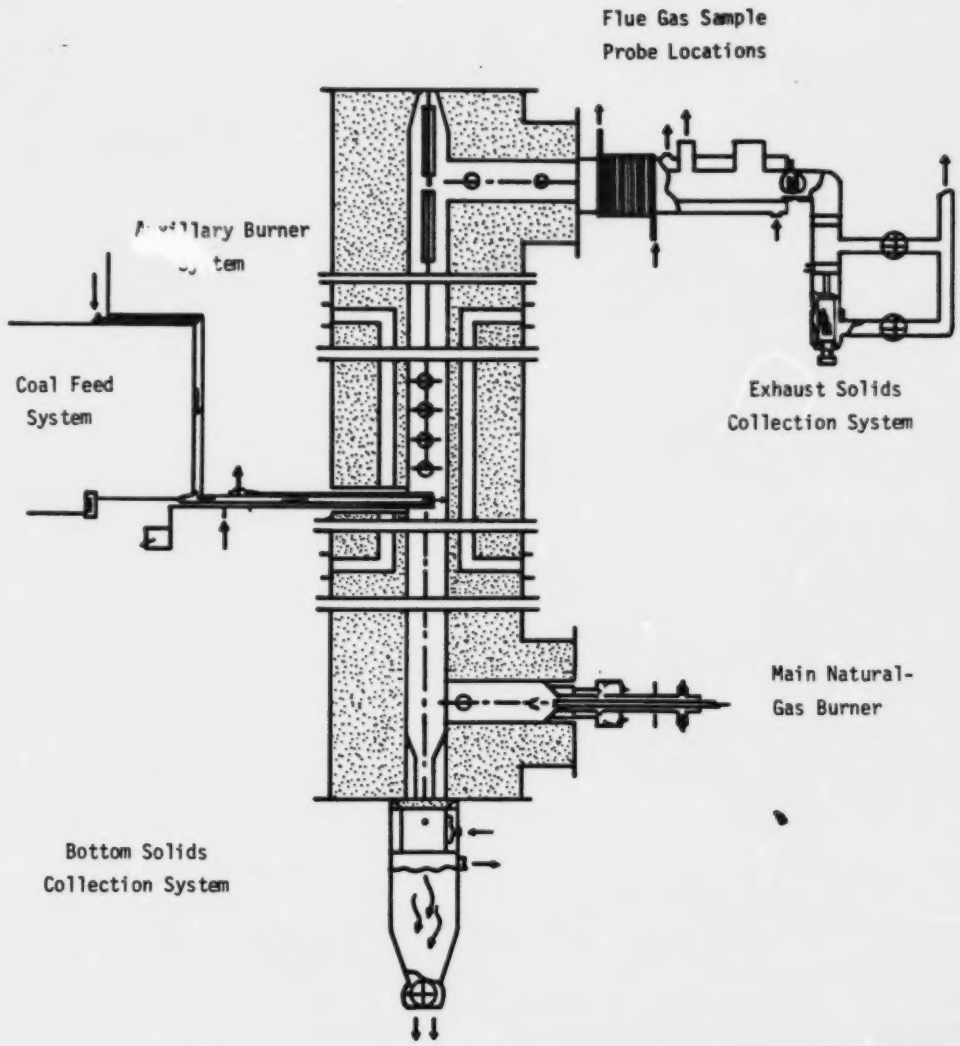

Figure 2.1. Tower Furnace Schematic

The coal particles were metered with a vibrating-bin, pulverized-coal feeder. The coal was then transported to a vibrating, water-cooled stainless steel injector located 32 inches from the bottom of the furnace. The coal particles delivered to the center of the furnace by the injector were either entrained or dropped. The particles falling to the furnace bottom were collected in a water-quenched trap containing six spray nozzles. Unburned particles reaching the flue were collected in a water-quenched, 200-mesh screen basket.

The overall experimental procedure was to determine the effect of specific combustion parameters on the evolution and oxidation of C, H, N, and S for a high volatile bituminous coal. The duration of an experimental run was approximately 30 minutes during which time the bulk gas was maintained at a constant temperature. During the first ten minutes of the run, the combustion products produced by the natural gas burner were analyzed to establish the gaseous species base-line (without coal). Ten minutes into the experiment, coal was injected into the furnace facility at a constant rate (approx. 2.0 lb/hr). This continued for the remaining 20 minutes of the experiment during which the total exhaust species were monitored. At the conclusion of the experimental run, the coal feed was terminated, and the gas species were allowed to return to base-line values.

The raw coal and solids obtained from either the bottom or exhaust collection systems were analyzed to determine elemental carbon, nitrogen, hydrogen and sulfur. The carbon, nitrogen, and hydrogen contents of the coal were determined by a Perkin-Elmer 240-B elemental analyzer, while a Fisher Model 475 sulfur analyzer determined the sulfur content. In the preparation of coal samples for analysis, ASTM standard procedures were followed.

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FIXED-BED FURNACE

The fixed-bed furnace was designed to provide a Lagrangian simulation of the time/temperature/environmental history experienced by small sections of the fuel-bed in a large, mass-burning traveling grate stoker. In this way the horizontal, planer dimensions were reduced dramatically compared to a full-size unit; however, the vertical dimension was scaled one-to-one to retain representative gas-phase residence times. Figure 2.2 shows a sideview of the fixed-bed furnace which was designed to have a maximum superficial firing rate of approximately 600,000 Btu/hrft^2. The furnace had outer dimensions of 27 inches by 23 inches by 102 inches and the inner combustion chamber was 15 inches by 11 inches in rectangular geometry.

The furnace was composed of four separate, flanged sections. Section A, the upper most section, contained a 6 inch circular flue and a 3 inch quartz window to provide an overhead view of the combustion chamber. Sections B and C were the central furnace pieces and contained overfire-air injection slots, large observation ports and probe ports. All three of the upper furnace sections had composite walls consisting of 1/8 inch carbon steel, 4 inches of block insulation and 2 inches of high-temperature refractory.

The lower furnace section, labeled D, contained the experimental batch-retort. The fuel-bed proper was 10 inches long, 6 inches wide and 6 inches deep with a total surface area of 0.42 ft^2. Immediately adjacent and surrounding the bed was a 1 inch high-temperature refractory wall which was used to contain the coal. The main natural gas burners were mounted on the batch-retort to provide uniform heating of the retort and combustion chamber prior to coal testing. The retort and gas burners were assembled in a

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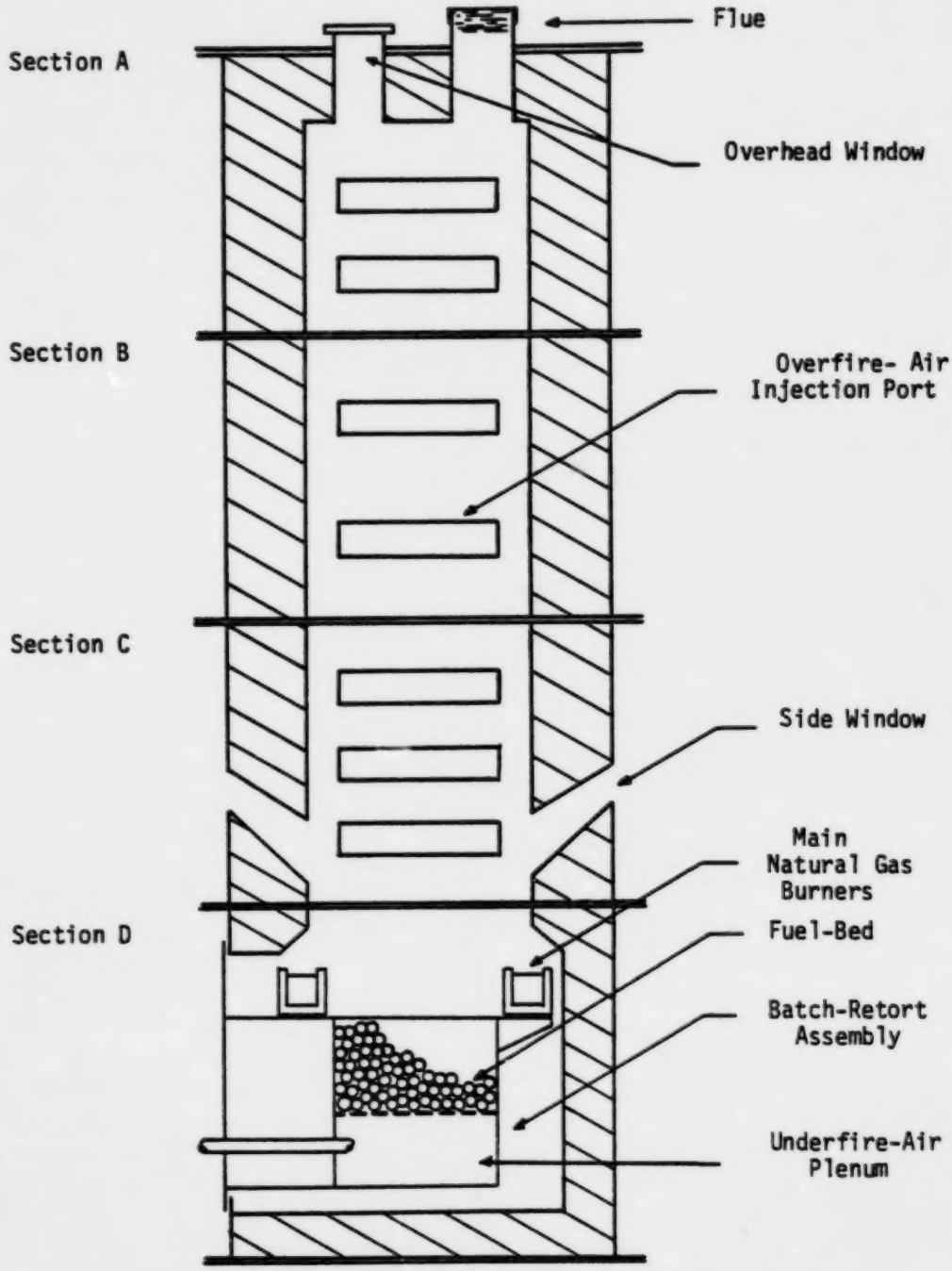

Figure 2-2. Sideview of the Fixed-Bed Furnace

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"drawer-like" configuration to permit easy loading of the coal particles before each experiment.

The combustion air was distributed between the underfire-air plenum in the batch-retort and two levels of overfire-air injection ports. The underfire air (primary combustion air) was supplied to a single plenum beneath the fuel-bed. The coal burned on a 1/4 inch thick stainless steel grate which contained 32, 5/16 inch holes spaced on 1.5 inch centers. The secondary combustion air was introduced through four overfire-air injectors each containing nine 9/64 inch holes, producing 100 ft/sec air jets. The overfire-air injectors were mounted on the furnace walls in pairs, opposing one another, to produce turbulent mixing. The overfire-air was evenly distributed between both pairs of injectors. All air streams were metered with rotameters but not preheated. Exhaust gas concentration measurements were made in the flue above Section A with a water-cooled stainless steel probe.

Prior to the coal experimentation, the fixed-bed furnace was brought to thermal equilibrium by firing natural gas for approximately nineteen (19) hours. The natural gas burners were then extinguished and the reactor drawer was withdrawn from the test section. Approximately eight pounds of coal were loaded into the bed and the charged reactor assembly was rolled back into the furnace and bolted into place. The combustion air was then adjusted and the coal bed usually ignited spontaneously within two minutes of insertion of the charged reactor drawer back into the furnace. If ignition was slow, the natural gas pilot was fired until the coal ignited, usually in less than 10 seconds, and then extinguished.

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MODEL SPREADER-STOKER FURNACE

The model stoker furnace (MSF) is shown in Figure 2-3. The furnace was designed to simulate the combustion environment found in a typical large-scale unit. Coal is delivered to the furnace by means of a calibrated auger-type feeder. The coal feed is deflected onto a rotating spreader which projects the coal particles into an overthrow region where small particles burn in the suspension phase. The spreader is located 32 inches above the grate and the total furnace height is 10.5 feet, thereby producing a vertical scaling of approximately 1:1 with typical field units.

The MSF was designed to fire coal at 60 lb/hr on a one ft^2 dumping grate, thus producing grate heat release rates which are comparable to full-scale boilers. The interior walls of the furnace consist entirely of high temperature castable refractories. The furnace is equipped with 600,000 Btu/hr natural gas burners which are used to preheat the combustion chamber to typical operating temperatures prior to experimental coal runs.

Air is injected into the furnace under the grate and at various heights above the grate. Air flow rates are measured with two laminar flow elements and the air is not preheated. The furnace chamber contains multiple access ports for insertion of sampling probes, internal cooling coils or for injection of overfire air.

ANALYTICAL METHODS

The combustion products from each of the furnaces were continuously monitored for NO, CO, CO_2 and SO_2. The analysis for SO_2 was accomplished by withdrawing a gas sample through an uncooled, quartz-lined, stainless steel probe and pumping it through a Dupont U.V. 400 Photometric Analyzer. Sample conditioning prior to the instrumentation consisted of two glass-wool filters,

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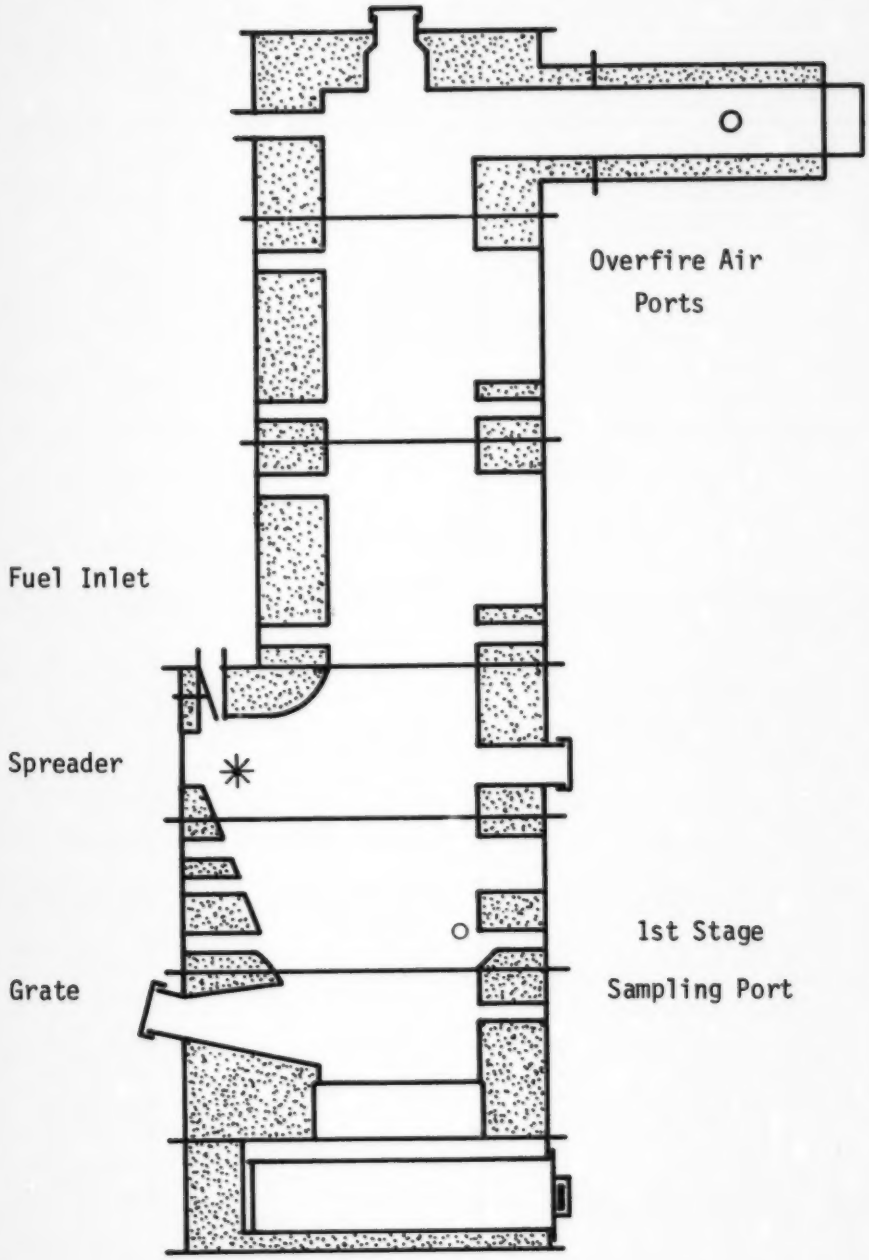

Figure 2-3 Model Stoker Furnace Cross-Section

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an Eaton electrically heated line (which maintained the sample at 250°F), two Purma-Pure dryers (which selectively removed water from the flue gas), and a 60-micron stainless steel filter.

To determine the other gas-phase species concentrations, combustion products were also drawn from the exhaust through a water-cooled, stainless steel probe and pumped into the instrumentation which included a chemiluminescent NO analyzer, a paramagnetic O_2 analyzer, and an NDIR analyzer for CO and CO_2. Sample conditioning prior to this instrumentation consisted of a refrigerated dryer, two glass-wool filters, a 7-micron stainless steel filter, and a stainless/Teflon sampling pump. All sample lines were a combination of 1/4-inch Teflon and stainless steel tubing with all fittings being 304 stainless steel.

FUEL ANALYSIS

Table 2-1 shows the ultimate and proximate analysis of each of the coals used in this study. The coals were sized for the tower furnace in narrow size ranges from 0.0 to 0.157"; the coals for the fixed bed furnace were sized at 1/4" to 1". In the model spreader stoker, the coals were burned on an as received basis.

Ultimate analysis data were obtained for all of the size ranges used in the suspension phase studies. These data indicated that both the total sulfur content and the sulfur forms varied with particle size. For the large coal particles the sulfate sulfur content was relatively small compared to the organic and pryitic sulfur content (as expected); however, as the coal particle size decreased, there was a corresponding increase in the sulfate sulfur content (Figure 2-4). In the smallest particle size range the sulfate sulfur content was of the same magnitude as the organic sulfur content.

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Table 2-1

Fuel Analysis

PROXIMATE ANALYSIS	UTAH	NORTH DAKOTA LIGNITE	INDIANA
Moisture	6.20	29.90	2.96
Ash	9.20	6.76	14.15
Volatile Matter	37.62	29.74	33.45
Fixed Carbon	46.98	33.60	49.44
Particle Size	1/4"- 1"	1/4" - 1"	0.0 - 0.063"

ULTIMATE ANALYSIS	UTAH	NORTH DAKOTA LIGNITE	INDIANA
Moisture	6.20	29.90	2.96
Carbon	66.93	44.74	61.79
Hydrogen	4.65	2.96	4.16
Nitrogen	1.11	0.67	1.24
Sulfur	0.33	0.37	2.17
Oxygen	11.58	14.60	13.53
Ash	9.20	6.76	14.15
Calorific Value (BTU/lbm)	11,571	7,573	10,829
Calorific Value (wet/mmf)[1] (BTU/lbm)	12,743	8,122	12,610
ASTM Rank[2]	HVC Bituminous	Lignite A	HVC Bituminous

Fuel Constituent (%, Daf[3])	UTAH	NORTH DAKOTA LIGNITE	INDIANA
Nitrogen	1.31	1.06	1.10
Sulfur	0.39	0.58	2.60

1 mmf: Mineral matter free
2 HV: high volatile
3 Daf: dry, ash free

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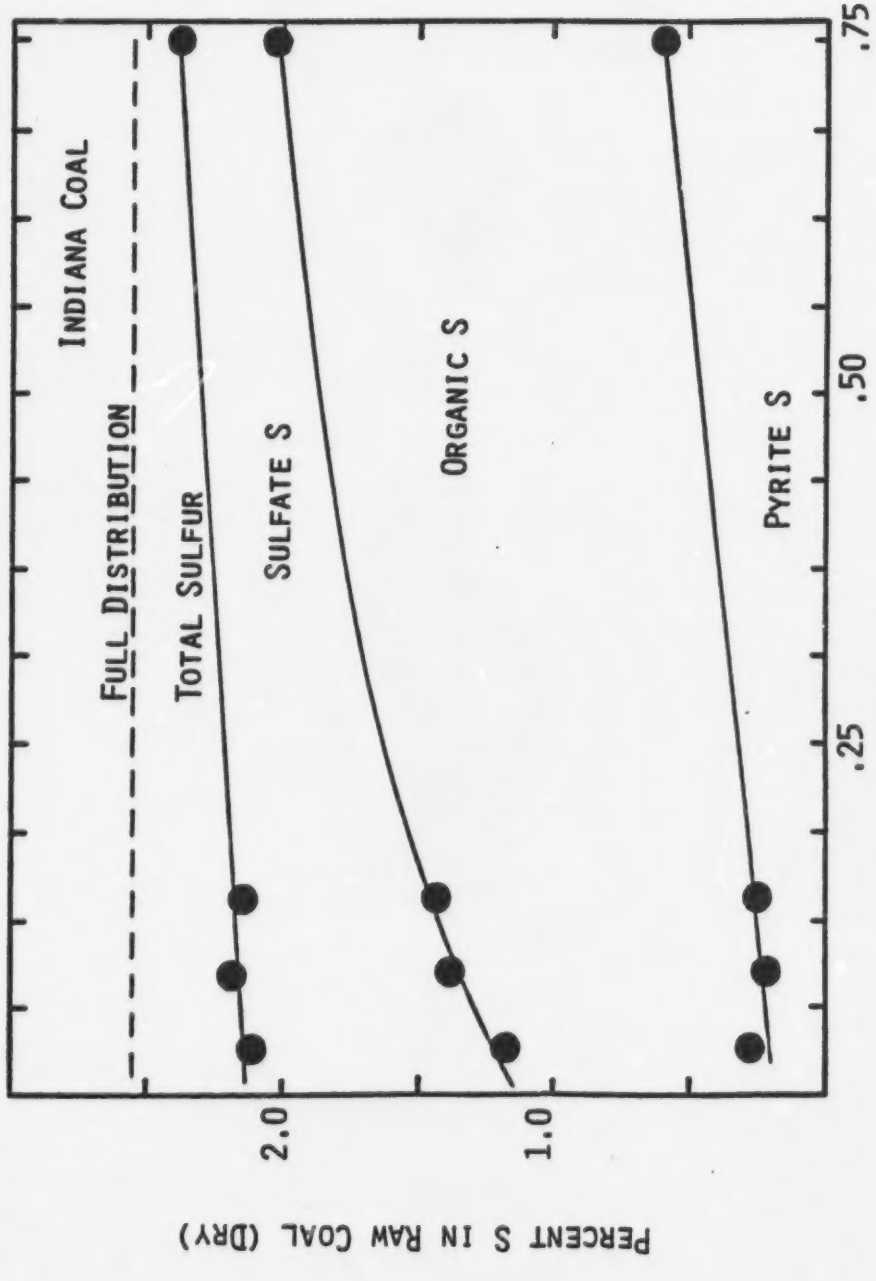

Figure 2-4. Influence of Raw Coal Particles Size on Distribution of Sulfur Forms

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This increase in sulfate sulfur may be partly attributed to a "weathering" effect. The "weathering" process begins when the coal is mined and continues until it is used. Weathering is defined as the oxidation of the sulfur (primarily inorganic pyrite) by the diffusion of water and air into the coal particles. Small particles weather much faster because of their higher surface-to-volume ratio.

However, the large increase in sulfate shown in Fig. 2-4 cannot be attributed only to weathering effects. The pyritic and sulfate sulfur content shown in Fig. 2-4 for the smallest mean particle size is larger than the available oxidizable inorganic sulfur, suggesting that the fines are broken off from larger particles at locations of high inorganic material concentrations (i.e., metal sulfates, pryitic sulfur, etc.). These locations would also correspond to the places where the mechanical strength of the coal structure is weakest. The increase of inorganic material due to breakage is supported by preliminary work performed with a scanning electron microscope where a high concentration of inorganic material was found at the fracture faces of the small raw coal particles under study. Figure 2-4, therefore, emphasizes the importance of using the "natural" coal fines and not a mechanically crushed coal when simulating the overthrow-phase combustion in a spreader stoker. In the suspension studies each particle range was obtained by mechanically screening the full distribution of stoker-sized coals.

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CHAPTER 3

SUSPENSION-PHASE COMBUSTION

LARGE PARTICLE STUDIES

The overall purpose of the suspension phase studies was to characterize the combustion in the overthrow region of a spreader stoker. The initial studies were conducted with the full particle size distribution (Figure 3-1) typical of commercial practice. The coal was metered with a 3" auger and was introduced at the top of the furnace through a water-cooled sleeve. The furnace was operated at 590,000 Btu/hr/ft^2, and the thermal environment was maintained at 1950°F with the combustion of full stoker-size coal particles. These data (Fig. 3-2a) indicated that increasing the oxygen concentration increased the amount of the coal burned; however, the small overall combustion percentages measured and visual observations suggested that only the smallest particles were combusted in the overthrow region.

To determine the extent of the large particle combustion in suspension, the large particle feeder was modified, as shown in Fig. 3-2b, to remove coal fines less than .1 inch from the coal feed. Figure 3-2c indicates the results when feeding only large coal particles. For two thermal conditions (1850°F and 1950°F) the large particle combustion in suspension was relatively small. At the higher thermal environment, increasing the oxygen concentration from 2 to 10 percent increased the coal burned from 2 to 2.5 percent. Thus, only the smallest coal particles (less than 0.1 inch) burn significantly in the suspension phase.

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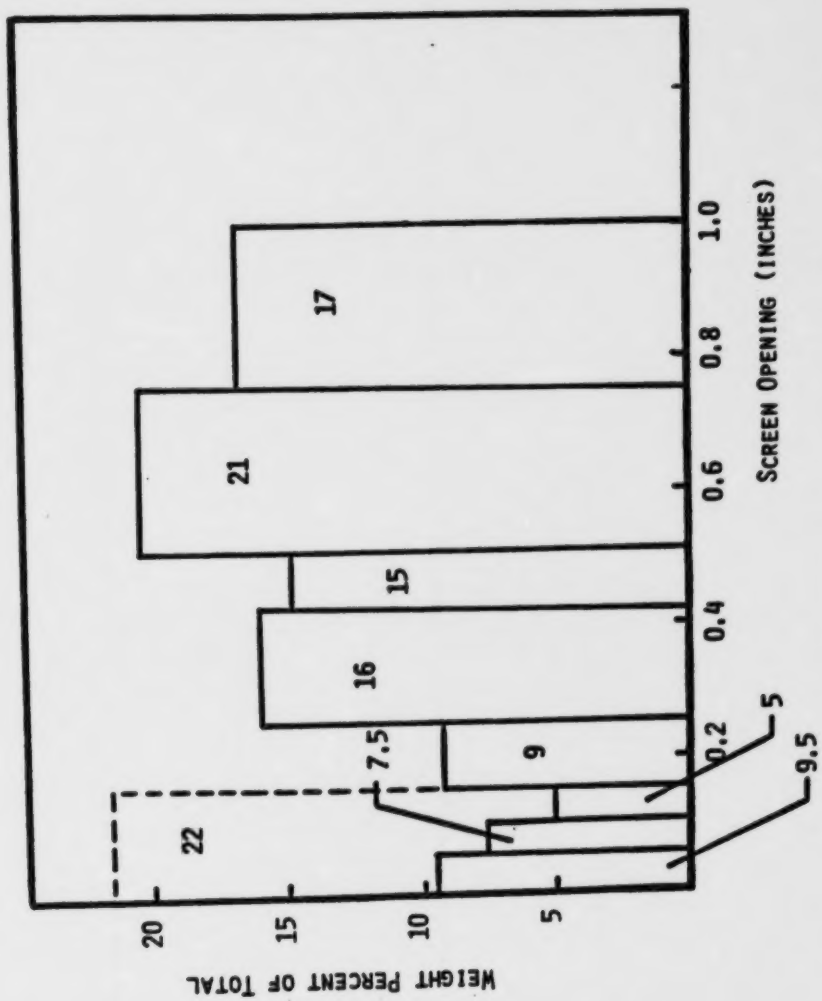

Figure 3-1. Stoker Coal Size Distribution

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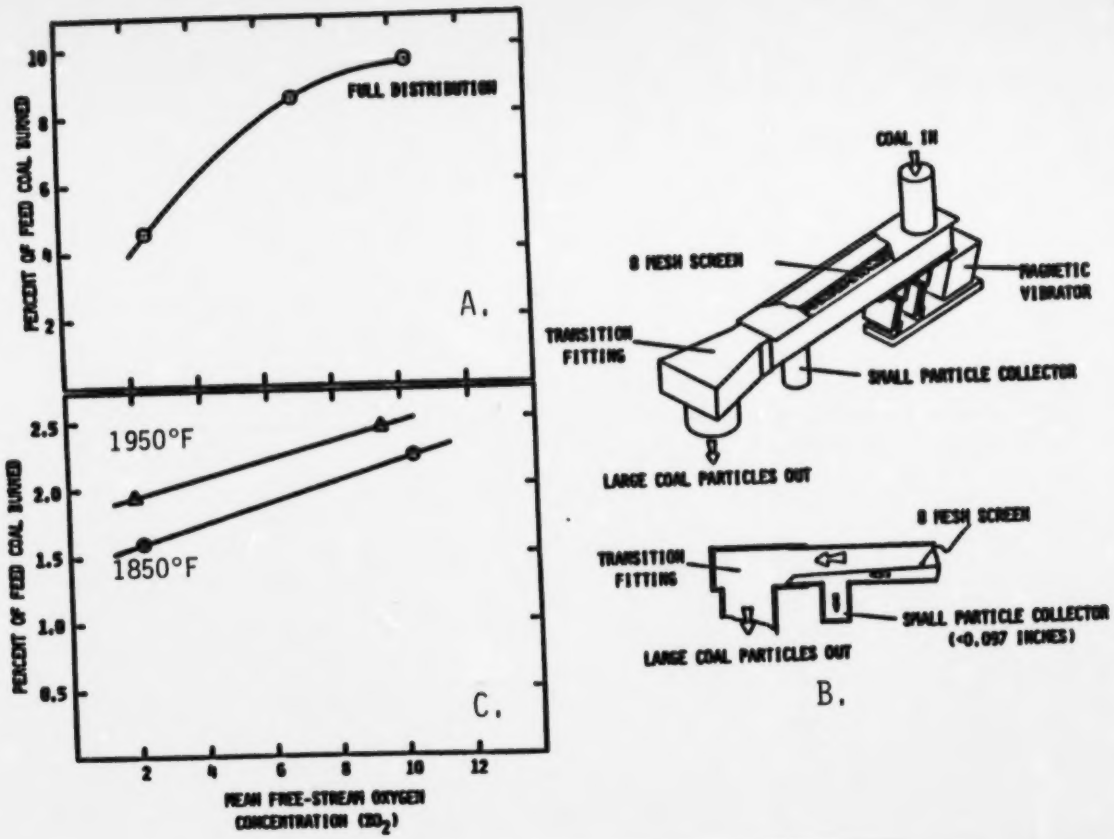

Figure 3-2. Large Particle Results

All subsequent suspension phase studies were concentrated on the three smallest particle size ranges (0.0 - 0.063, .063 - .111, .111 - .157 inches) shown in Figure 3-1. These size ranges account for approximately 9.5, 7.5, and 5.0 percent of the total weight of stoker coal, respectively. The three coal-particle ranges together encompass approximately 22 percent of the coal mass delivered into a typical industrial spreader-stoker.

FATE OF MAJOR CONSTITUENTS

Figure 3-3, 3-4, and 3-5 present results typical of those obtained in suspension furnace. These data were gathered by analyzing the gas phase for CO, CO_2, NO, and SO_2 and by determining the mass and composition of the solid materials collected at the furnace top and bottom. (H_2O was estimated by a differential hydrogen balance). The data shown are for the 1900°F thermal condition (wall temperature = 1900°F), the Indiana coal, the smallest 10 percent of a commercial stoker feed (d < .063 inches), and a natural gas firing rate equivalent to a bed-heat release rate of 670,000 Btu/hr/ft^2. (Commercial practice ranges from 500,000 to 750,000 Btu/hr/ft^2.)

Carbon

Figure 3-3 indicates that for the smallest particle size range, more than 50 percent of the inlet carbon was oxidized at all conditions. The data are plotted in terms of mean free-stream oxygen which is the average of the oxygen concentration at the top, and the bottom of the furnace. (In general, the range was approximately ± 1% O_2.) Increasing the mean free-stream oxygen decreased the material collected at the furnace bottom (the particles which would fall to the grate in a commercial system) because of increased entrainment. Increasing the excess oxygen from 2 to 10 percent increased the vertical velocity from 11 to 19 feet per second. CO_2 formation increased with

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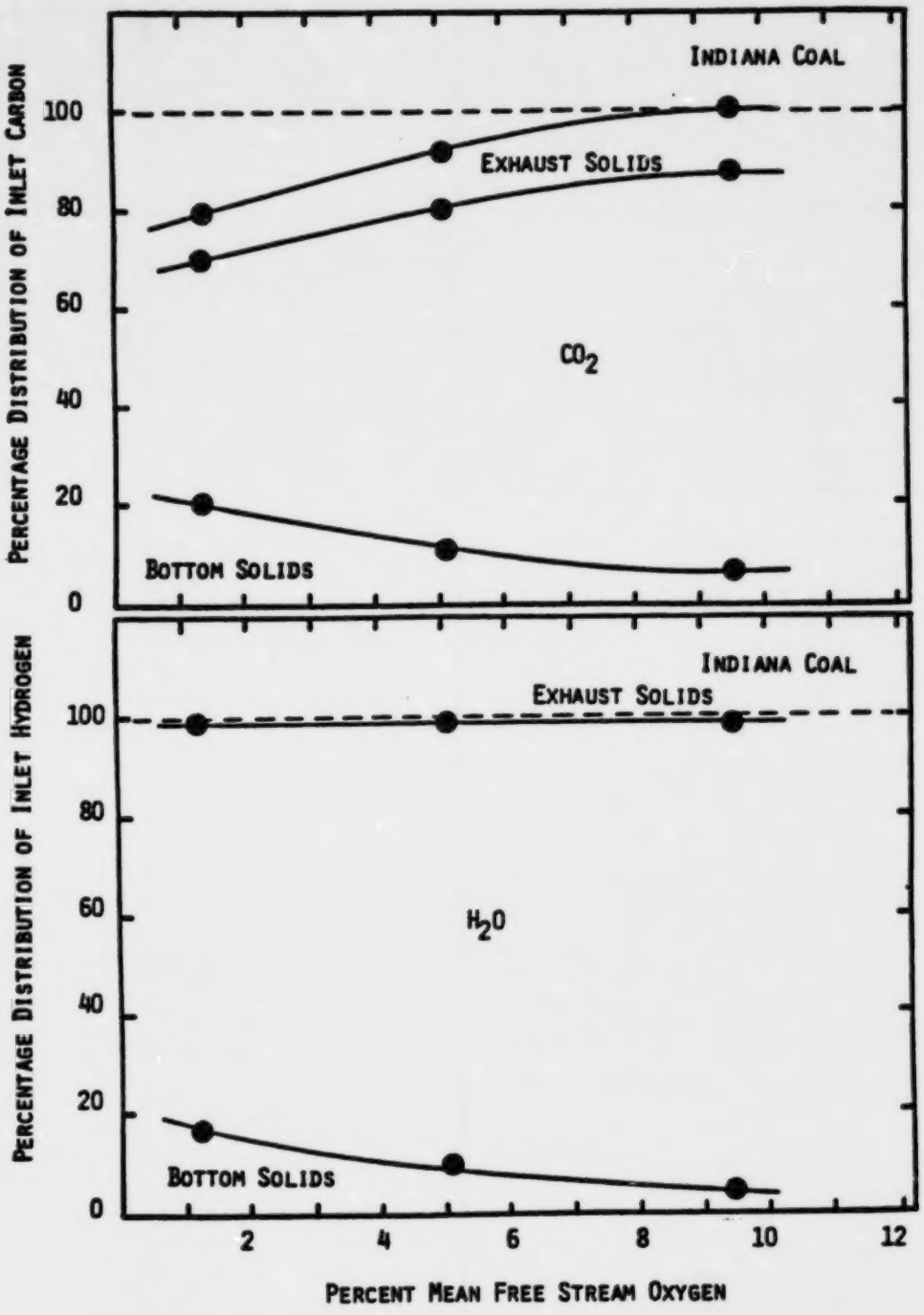

Figure 3-3. Fate of Carbon and Hydrogen in the Tower Furnace for Particles <.063" with 1900°F Thermal Environment and 670,000 Btu/hr/ft² SBHRR

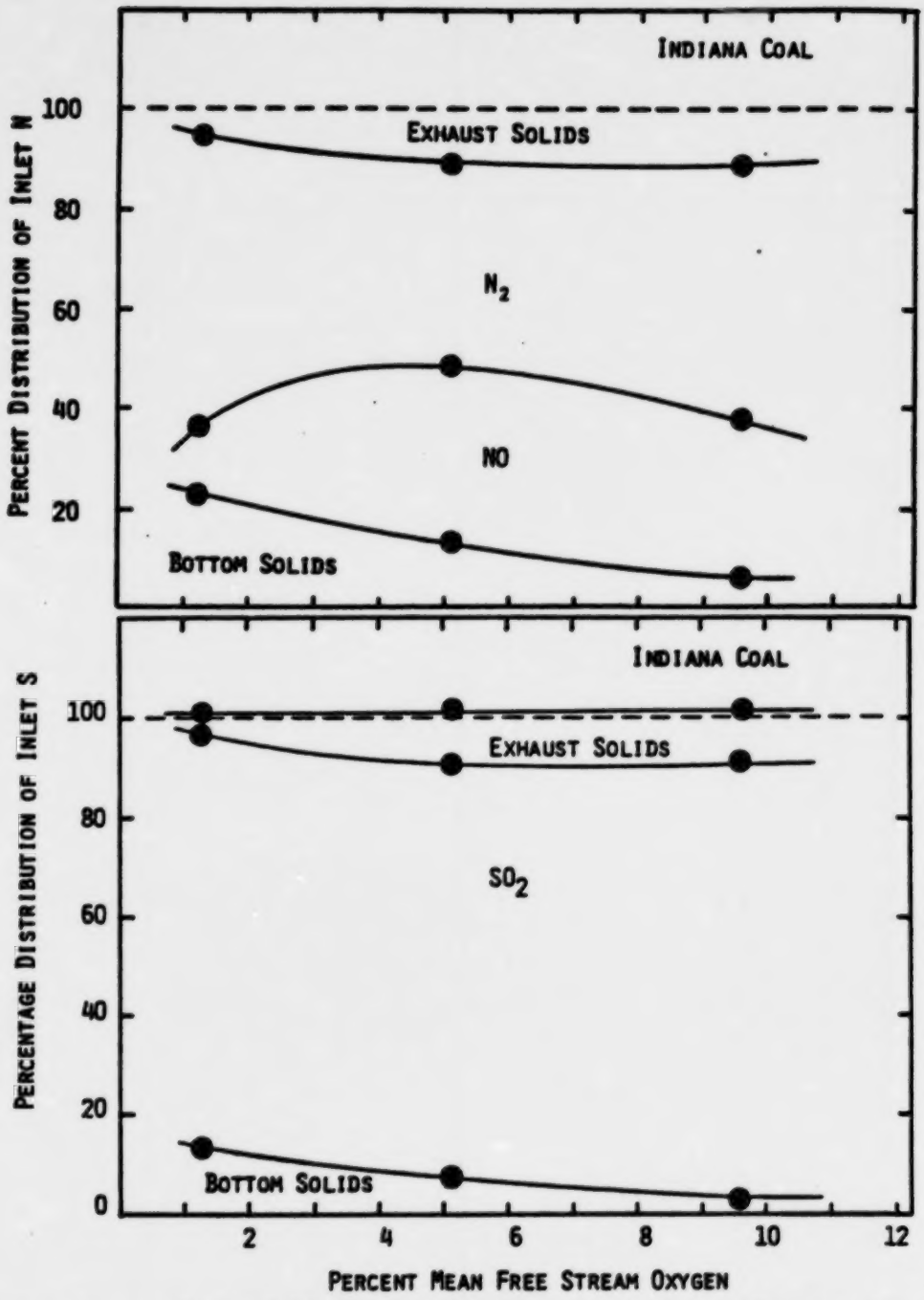

Figure 3-4. Fate of Nitrogen and Sulfur in the Tower Furnace for Particles <.063 with 1900°F Thermal Environment and 670,000 Btu/hr/ft^2 SBHRR

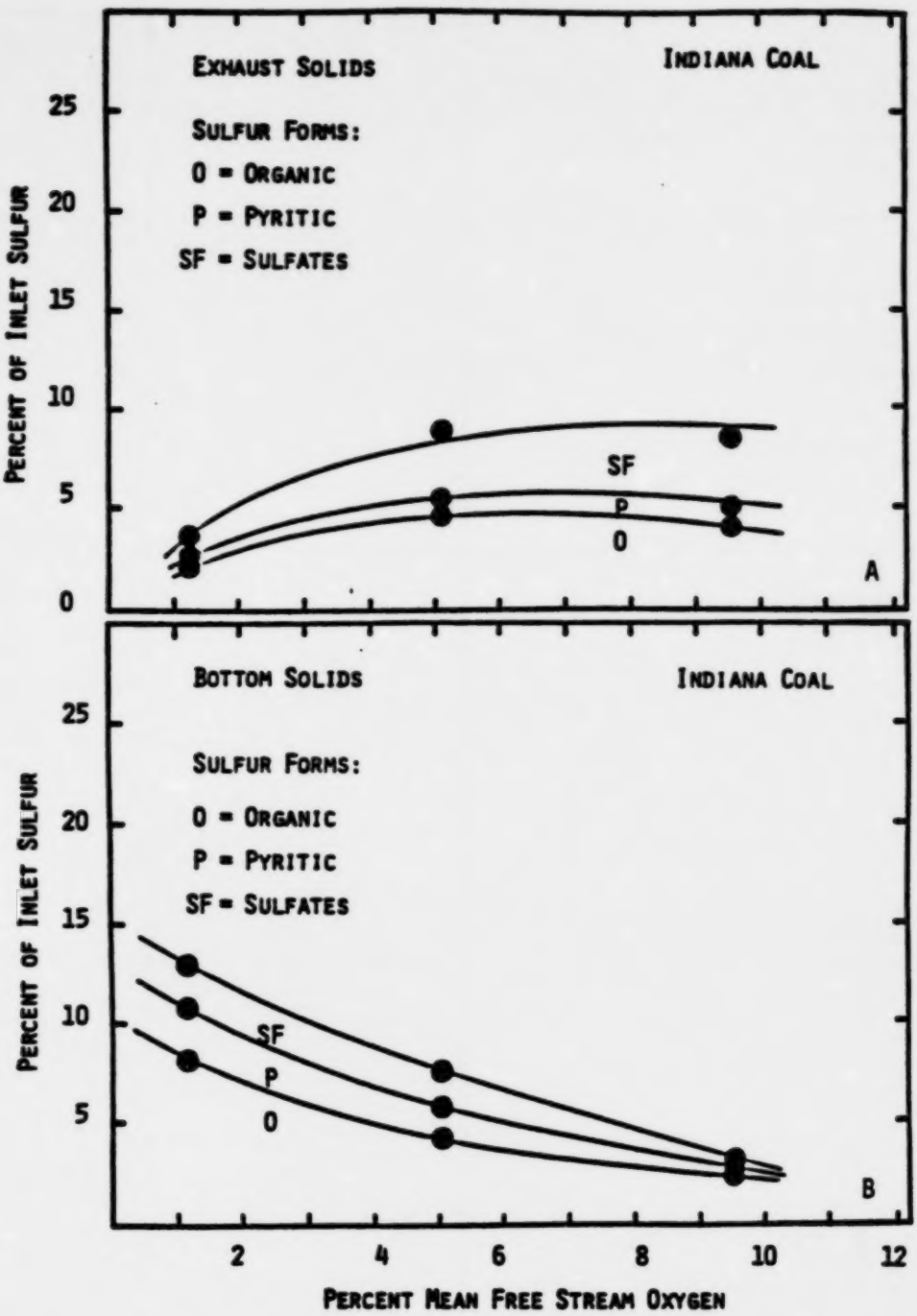

Figure 3-5. Distribution of Sulfur Forms in Exhaust and Bottom Solids for Coal Particles <.063 inches and 1900°F Thermal Environment

increasing oxygen due to both increasing entrainment and increasing oxygen partial pressure. Both effects must have been significant because the increase in CO_2 greatly exceeded the decrease in bottom carbon (Fig. 3-3).

The overall carbon balance was excellent at high mean free-stream oxygen levels, but at low oxygen concentrations approximately 20 percent of the carbon could not be directly accounted for. This deficiency was believed to be primarily due to the formation of carbonaceous soot which was not collected by the 200-mesh (88 microns) screen system. Visual observation indicated that large amounts of soot were formed at the low oxygen condition.

Hydrogen

Figure 3-3 shows similar data on the fate of hydrogen. The hydrogen content of the bottom solids decreased with increasing mean free-stream oxygen. This trend closely parallels the carbon behavior, but the absolute magnitude was slightly lower, indicating that some hydrogen is evolved from the particles which fall to the grate. The exhaust solids contained essentially no hydrogen, suggesting that the material which is carried over head in a commercial unit is primarily devolatilized char. It is, therefore, not surprising that this material often proves difficult to burn, even in a furnace equipped with a "fly-ash reinjection system."

Nitrogen

Figure 3-4 shows data on the fate of nitrogen. NO formation was determined by measuring the NO concentrations with and without the coal feed. (N_2 was determined by difference.) Increasing the mean oxygen decreased the fraction of nitrogen reaching the bed and slightly increased that carried into the exhaust; both effects are primarily due to increased particle entrainment. NO formation decreased significantly at low mean free

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stream oxygen concentrations as expected from pulverized coal data.[14] Particles which fell to the bottom, burned only slightly (hydrogen concentrations still significant) and exhibited some nitrogen enrichment. Thus, the evolution of nitrogen slightly lagged the evolution and oxidation of the early hydrocarbon volatiles and this is in agreement with inert pyrolysis studies.[6, 15] However, the exhaust solids were deficient in nitrogen compared to the raw coal, demonstrating that the final nitrogen compounds reacted preferentially to the most refractory char species. Since the exhaust solids exhibited behavior opposite that of the bottom solids, and since their relative masses were approximately equal for the smallest particles (<.063 inches), these data indicate that the overall nitrogen evolution rate was near that of carbon.

The fate of the total sulfur is also shown in Fig. 3-4. In general, the sulfur balance closure was significantly better than that achieved with carbon, probably because the uncollected soot (the problem with the carbon balance) contained little sulfur. Overall, sulfur was found to be less reactive than hydrogen, but more reactive than carbon and nitrogen, which agrees with previous inert pyrolysis data.[6, 15] The sulfur content of the bottom solids decreased with increasing mean free-stream oxygen and again this was believed to be the direct result of increased entrainment due to the increased vertical velocity. The SO_2 formation remained surprisingly constant as the oxygen concentration was increased because the sulfur content of the exhaust solids increased. To understand this behavior it is necessary to consider the fate of the individual sulfur components.

Figure 3-5 shows the detailed distribution of the sulfur forms in both the bottom and exhaust (top) solids. All three sulfur forms in the bottom

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solids decreased with increasing mean free-stream oxygen due to the increased entrainment. In the raw Indiana coal, the organic sulfur and the sulfate sulfur were approximately equal (Table 1); however, in the bottom solids, the sulfate sulfur was significantly less than the organic sulfur.

Figure 3-5 also indicates that all forms of the sulfur in the exhaust solids increased with increasing mean free-stream oxygen; however, the increase in the sulfate sulfur was unexpectedly dramatic. Increasing the oxygen concentration from 2 to 10 percent increased the exhaust carbon by 100 percent and the organic sulfur by 125 percent. The similarity of these percentages suggests that organic sulfur follows the evolution of carbon as expected. However, the sulfate sulfur increased by more than 230 percent in this case. This large change in sulfate sulfur is reasonable because sulfate components are particularly easily decomposed at low oxygen levels; equilibrium calculations for metal sulfates suggest that the 50 percent decomposition temperature drops sharply as the local oxygen concentration is reduced to near zero.

INFLUENCE OF PROCESS VARIABLES

A series of experiments was conducted to establish the influence of mean free-stream oxygen concentration, particle size, and thermal environment on the fate of carbon, hydrogen, nitrogen, and sulfur in the suspension phase. Reactor wall temperatures of 1900°F and 2100°F were studied. The wall temperature in the main reaction zone was measured with an optical pyrometer and maintained within ± 25°F of the set point. Particle size was varied by mechanically screening the raw coal into narrow size ranges, each corresponding to approximately 10 percent of the normal spreader-stoker size distribution.

Carbon

Figure 3-6 summarizes the influence of the combustion parameters on the fraction of the feed carbon oxidized to CO_2 in the suspension phase. Increasingly mean free-stream oxygen from 2 to 10 percent increased the carbon oxidized at all combustion conditions studied. As discussed previously, this was due to the five-fold increase in oxygen partial pressure and to the increased entrainment caused by the higher vertical velocity.

Particle size had a dramatic effect on CO_2 formation. At the nominal commercial operating conditions (6% O_2, 2100°F), approximately 80 percent of the coal particles less than .063 inches burned in the suspension phase, while less than 20 percent of the particles between .063 and .111 inches burned. Thus, particles small enough to be entrained in the upward flow oxidized almost completely; but large particles which fell directly to the grate remained essentially unreacted.

Increasing the reactor temperature by approximately 200°F increased the combustion of the entrained particles due to both increased devolatilization rates and increased reaction rates. Little effect of bulk temperature was observed at low O_2 concentrations and large particle sizes because essentially no particles were entrained at this condition. Simple drag calculations, assuming spherical particles with constant density, indicated that at the 2 percent oxygen condition, particles greater than .025 inches would fall, while at the 10 percent oxygen condition, particles up to .035 inches would be entrained.

Hydrogen

Figure 3-6 shows that, in general, the influence of the combustion parameters on hydrogen oxidation is weaker than on carbon, probably because

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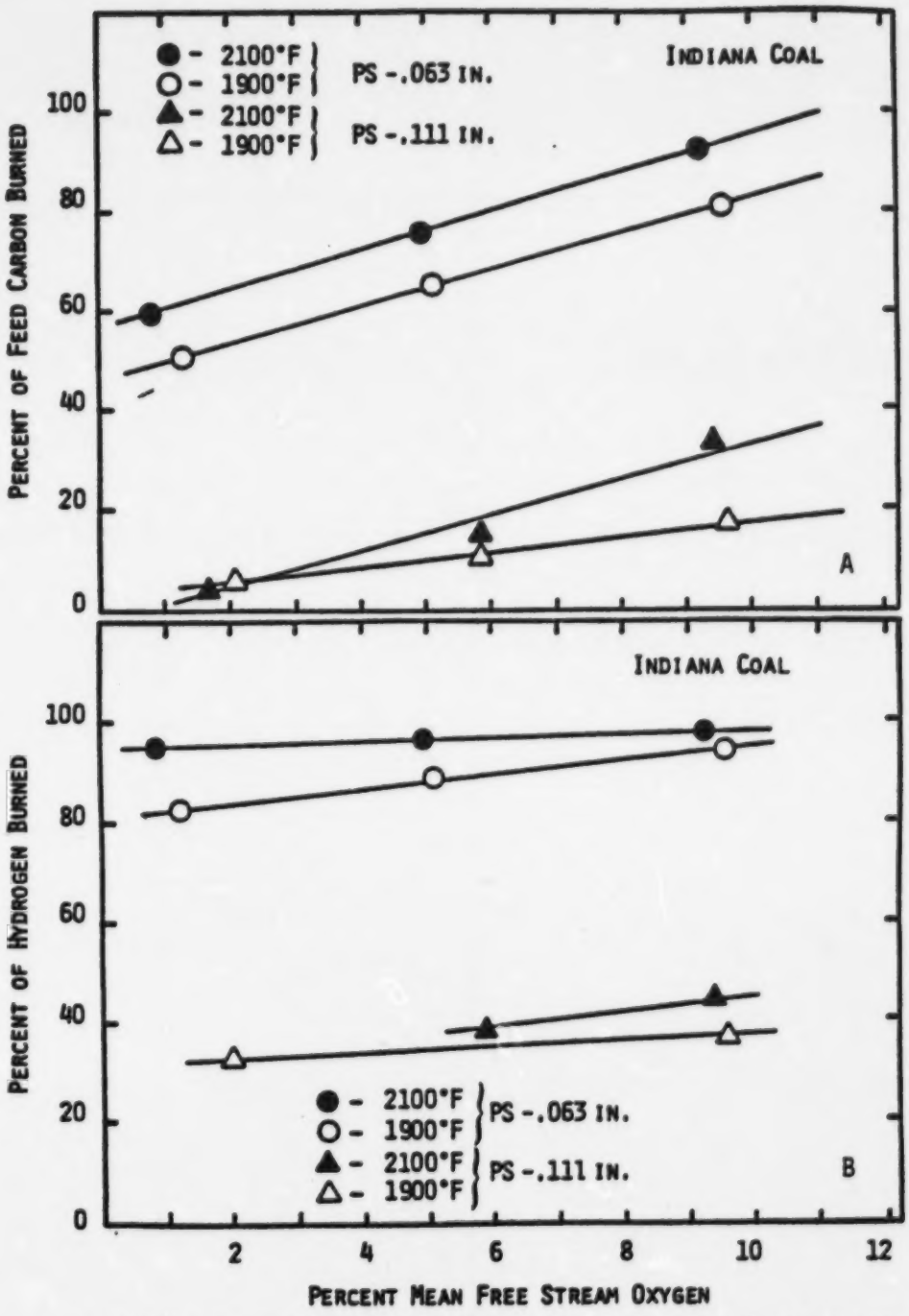

Figure 3-6. Influence of Combustion Parameters, Thermal Environment, Oxygen Partial Pressure and Coal Particle Size on Carbon and Hydrogen Evolution

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volatile evolution is almost complete with all particles which are entrained. Increasing the reactor temperature by 200°F slightly increased the hydrogen oxidation due to the increased volatile evolution rates. Increasing the mean particle size dramatically reduced the hydrogen oxidation because of reduced entrainment; however, even with the large particles, hydrogen oxidation exceeded carbon oxidation at all conditions.

Nitrogen

Figure 3-7 summarizes the effects of the combustion parameters on the percent of the evolved fuel nitrogen which was ultimately converted to NO. Conversions were relatively high (c.a. 40%) and essentially independent of the mean free-stream oxygen, except at very low O_2 levels. The smaller particles exhibited slightly higher fuel nitrogen conversions (Fig. 3-7a) and this agrees with pulverized coal results on much smaller particles.[16] Decreasing particle size is known to increase the volatile nitrogen evolution rate and believed to enhance the availability of O_2 for XN oxidation. (The importance of intra-particle XN reduction reactions is decreased as particle size decreases.)[17] Bulk gas temperature had relatively little effect on fuel nitrogen conversion (Fig. 3-7b).

Figure 3-8 shows the nitrogen retained in the solid phase relative to the solid phase carbon in the char (normalized with respect to nitrogen to carbon ratio in the unburned coal) obtained from the bottom and top (exhaust) solids collectors. Included in Fig. 3-8 are the analysis results for two thermal conditions and also two different particle sizes. Increasing the bulk gas temperature by 200°F had little influence on the nitrogen-to-carbon ratio of the bottom solids for the smallest particle (<.063 inches), however, the exhaust solids N/C ratios were consistently lower at the higher temperature

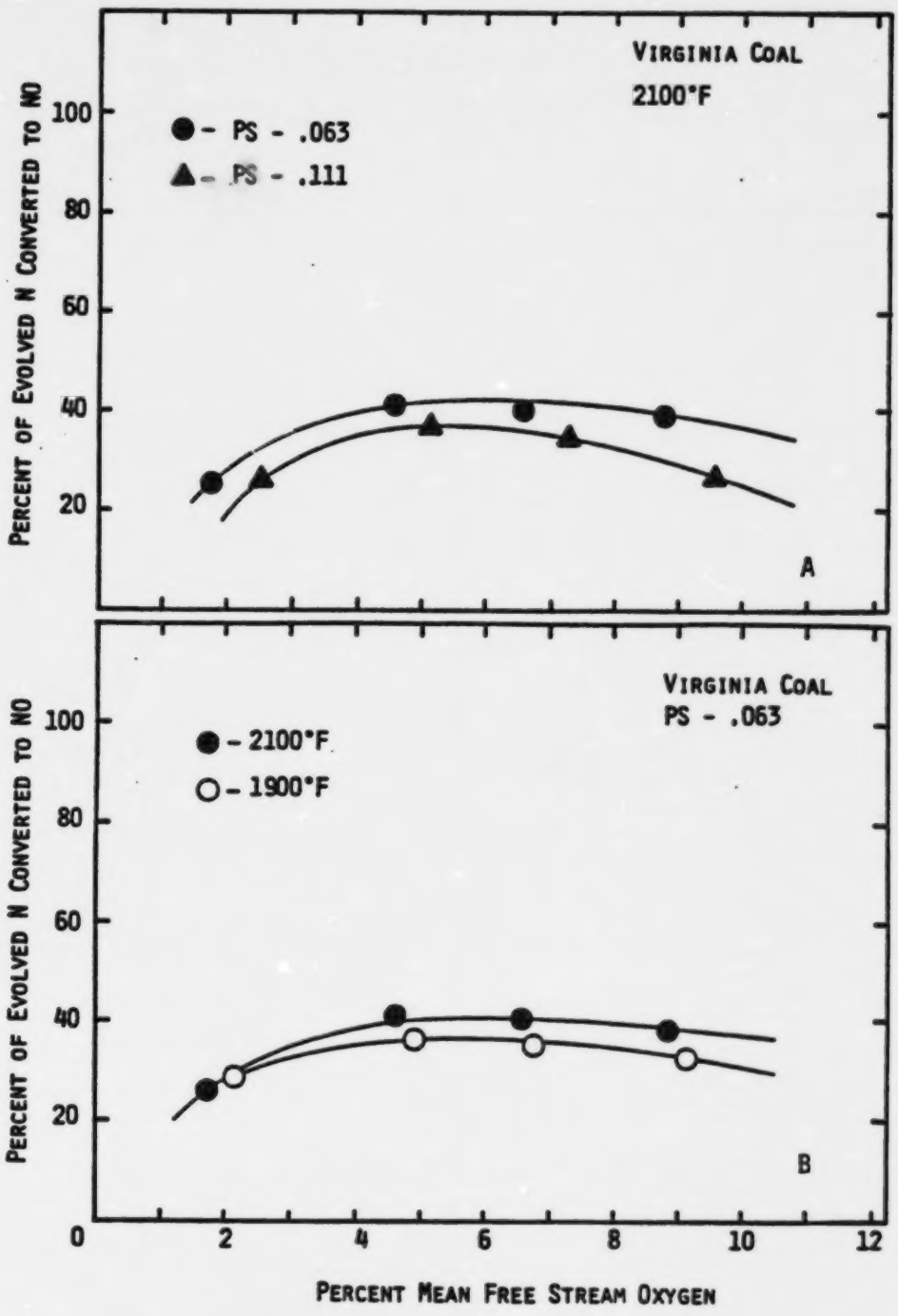

Figure 3-7. Influence of Combustion Parameters-Thermal Environment, Oxygen Partial Pressure, and Particle Size on Evolution of Nitrogen.

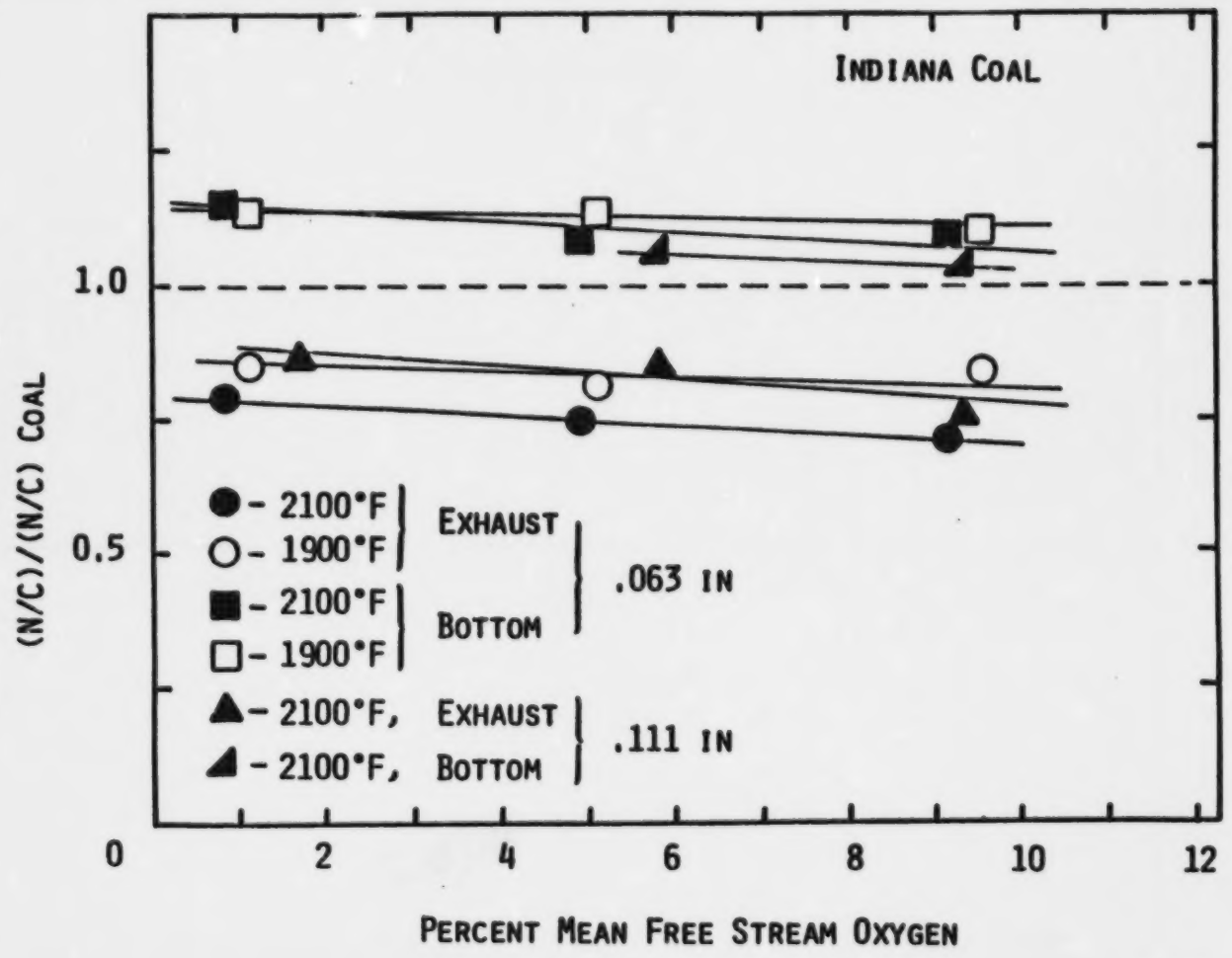

Figure 3-8. Solid Nitrogen Analysis for Material Obtained in Bottom and Exhaust Solid Collectors after Combustion of Particles.

which suggested that increasing the bulk gas temperature increased fuel nitrogen evolution relative to carbon oxidation.

With the larger particles, the exhaust solids showed a significant decrease in the magnitude of preferential evolution of the nitrogen from the most refractory char species. These results indicated that decreasing particle size, increased fuel nitrogen evolution. With relative masses considered, the results indicate, in general, that the overall nitrogen evolution rate was near that of carbon for the size of particles studied.

Sulfur

Figure 3-9 shows the influence of the combustion parameters on the oxidation of total sulfur. Again, there was a strong influence of particle size due to entrainment effects.

In general, increasing temperature increased the sulfur evolution and oxidation even more than carbon oxidation. Further, the dependence on mean free-stream oxygen was exactly opposite that observed with carbon (Figure 3-5); with the smallest particles increasing oxygen had little effect on sulfur oxidation; but with the larger particles, increasing oxygen decreased sulfur oxidation, particularly at the high temperature. Solids analysis data (like that shown in Fig. 3-5a for the Indiana coal) suggested that this difference was due partly to the reduced stability of the metal sulfates at low oxygen concentrations. Thus, the amount of the organic sulfur which is oxidized depends primarily on the overall coal oxidation rate, while the rate of inorganic sulfur decomposition depends primarily on the local temperature and the local oxygen concentrations.

Figure 3-10 further emphasizes the importance of this effect by presenting these results in terms of the dimensionless SO_2/CO_2 ratio

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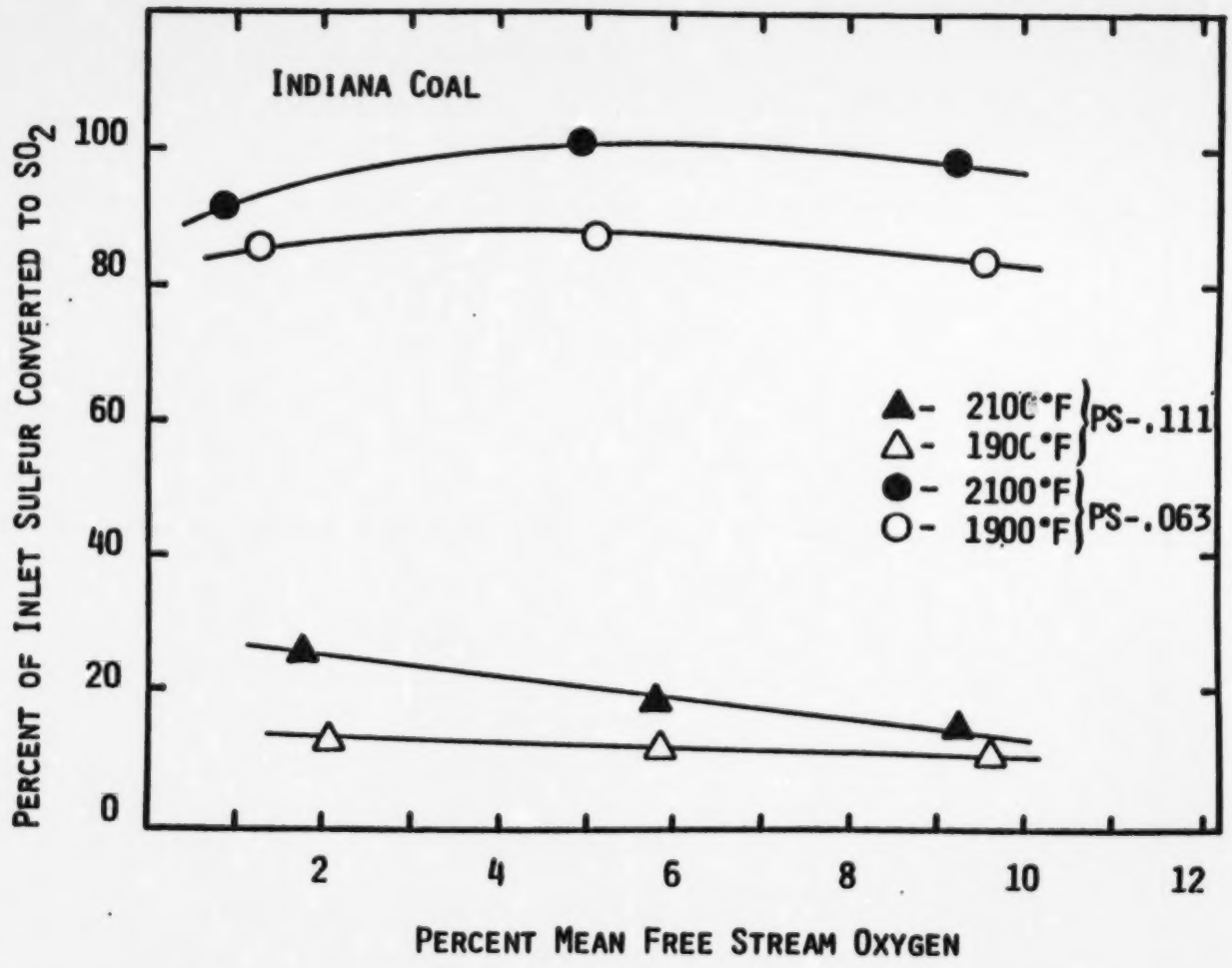

Figure 3-9. Influence of Combustion Parameters--Thermal Environment, Oxygen Partial Pressure and Particle Size on Oxidation of Total Sulfur

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normalized with respect to the sulfur-to-carbon content of the original coal. This figure includes the data for both thermal conditions and two particle sizes. When the oxygen availability is limited (low oxygen partial pressures), sulfur will be oxidized preferentially to carbon, and this effect can be relatively large (two fold); but, when oxygen is in great excess, the carbon oxidation may even lead sulfur oxidation slightly.

Figure 3-11 shows the detailed solid analysis results for the two thermal conditions with the smallest particles (<.063 inches). These data are reported in terms of the sulfur-to-carbon ratio in the collected solid relative to the sulfur-to-carbon ratio in the raw coal. The upper portion of Fig. 3-11 indicates that, in general, sulfur oxidation exceeds carbon oxidation (ratios less than 1.0). Increasing the bulk temperature by 200°F had little influence on the sulfur to carbon ratio in the bottom solids; however, it consistently decreased the sulfur-to-carbon ratio in the exhaust solids (i.e., high temperatures enhance sulfur evolution and oxidation). Figure 3-11b (lower graph) shows the ratio for the organic components of the sulfur only. These data are much closer to 1.0 and again indicate that, as expected, the organic sulfur follows the carbon. (It increased both the carbon and organic sulfur evolution by the same amount.) Thus, since increasing temperature significantly reduced the relative total sulfur content of the exhaust solids (Fig. 3-11a) but had little effect on the organic sulfur (Fig. 3-11b), it must have significantly increased the decomposition of the sulfate sulfur.

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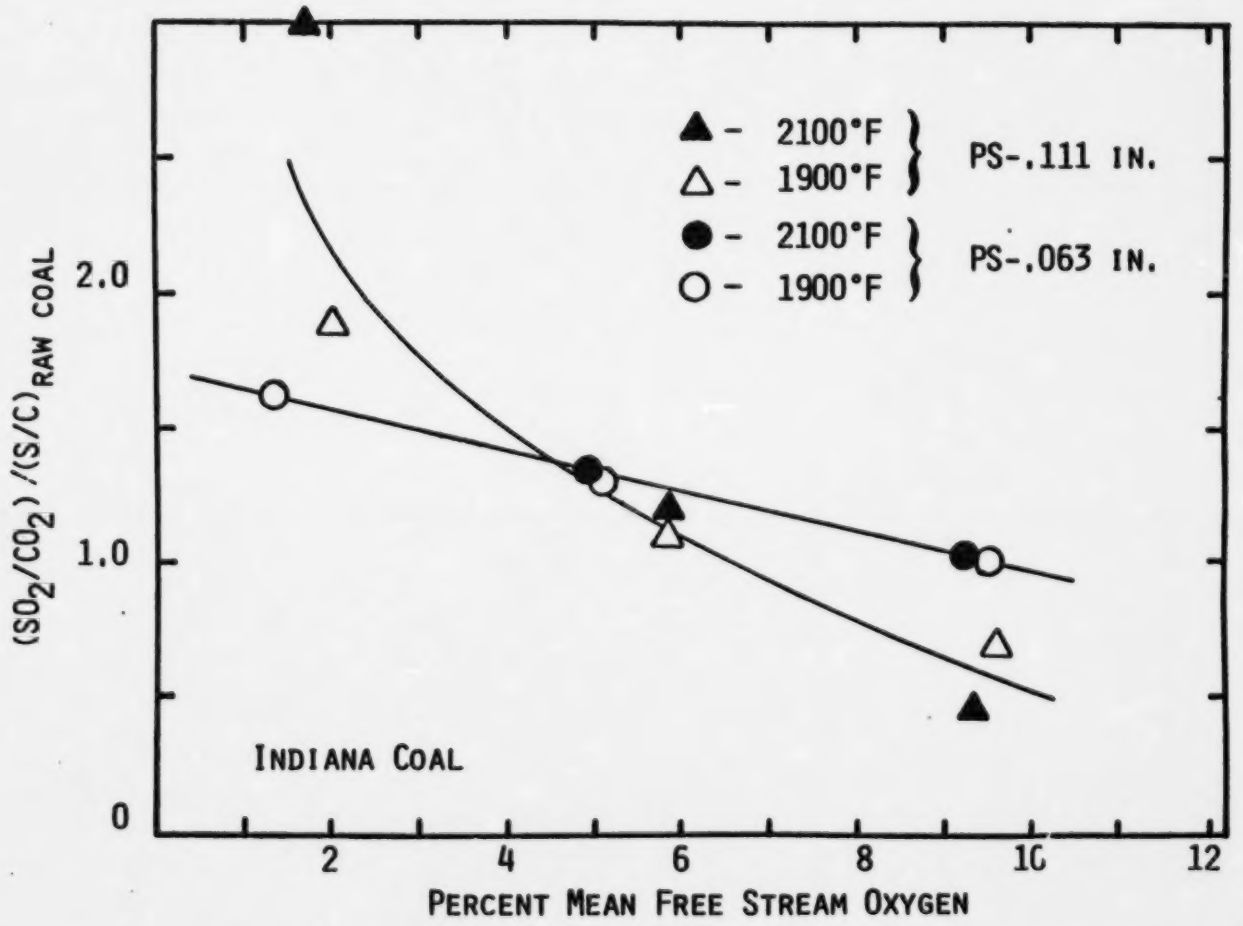

Figure 3-10. **Influence of Oxygen Partial Pressure on Sulfur and Carbon Oxidation.**

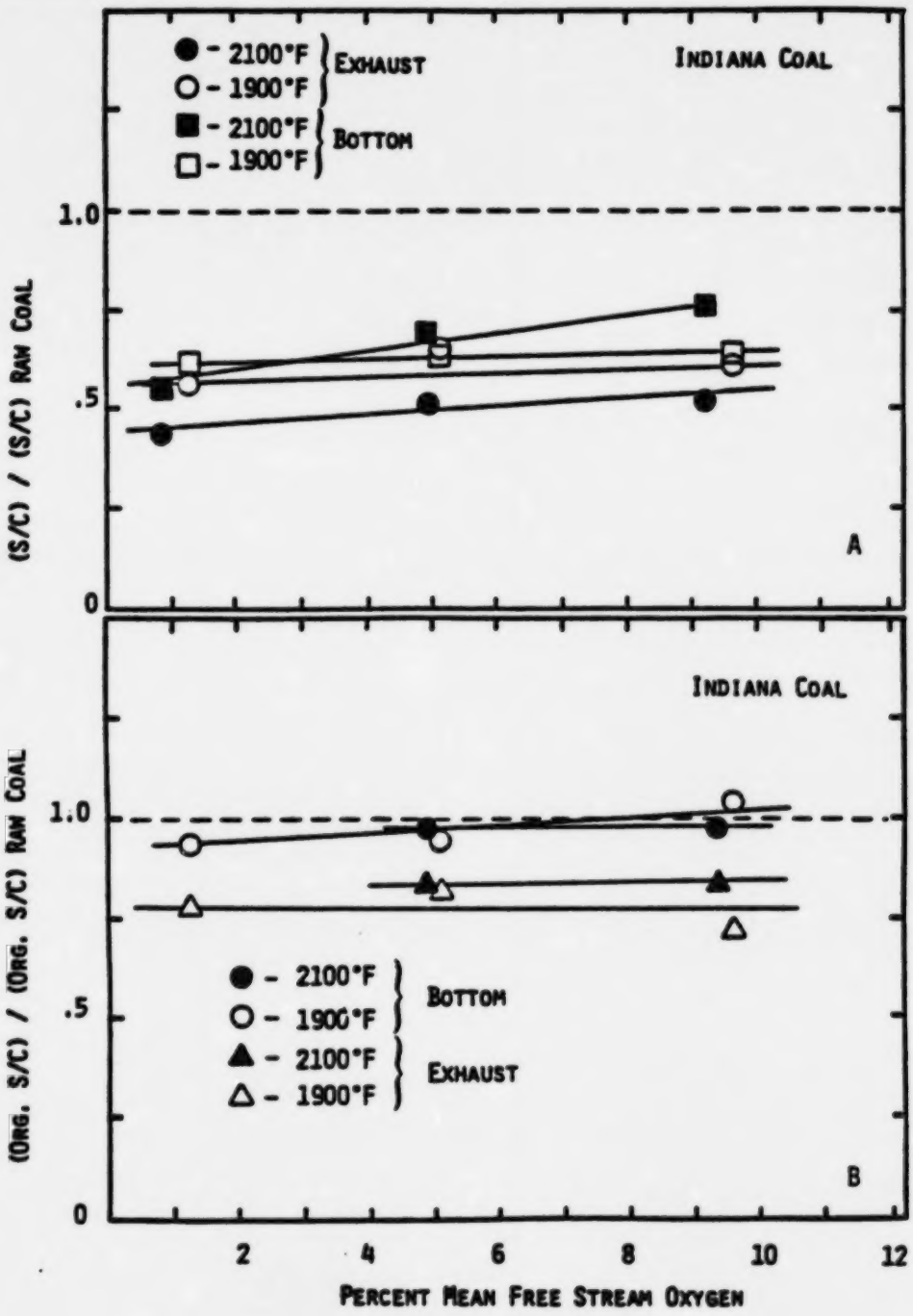

Figure 3-11. Solid Sulfur Analysis for Material Obtained in Bottom and Exhaust Solid Collectors After Combustion of Particles <.063 in.

CONCLUSIONS

Hydrogen Oxidation

The results of this study indicate that particles greater than 0.1 inches reach the bed of a stoker essentially unreacted, and particles less than 0.06 inches burn completely in the suspension phase. Hydrogen evolution and oxidation significantly leads overall carbon oxidation so that particles carried into the exhaust are completely devolatilized. Increasing excess oxygen enhances suspension phase burning due to increased particle entrainment and increased oxygen partial pressure. Increasing the bulk gas temperature also increases both the carbon and hydrogen oxidation.

Nitrogen Oxidation

Fuel nitrogen is evolved at approximately the same rate as carbon is oxidized. The conversion of the evolved nitrogen is relatively high (c.a. 40%) and depends primarily on local oxygen availability. Particle size and thermal environment effects are of second order importance.

Sulfur Oxidation

The initial sulfur distribution in the raw coal varies with particle size; naturally occurring fines are unusually high in sulfate sulfur. The evolution and oxidation of organic sulfur appears to depend primarily on the rate of carbon evolution; however, the rate of sulfate sulfur decomposition is most strongly influenced by local oxygen concentration and temperature. At low oxygen levels, sulfur evolution significantly leads carbon oxidation; and this effect appears to be directly related to the decreased thermal stability of metal sulfates at low oxygen partial pressures. The initial sulfur distribution does not strongly influence the overall SO_2 formation.

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CHAPTER 4

FIXED-BED STUDIES

TYPICAL EXPERIMENTAL RESULTS

The purpose of the fixed-bed experiments was to characterize the evolution of fuel nitrogen and sulfur as a function of time for a thick fuel bed typical of commercial chain and traveling grate stokers. All of these tests were conducted with the Utah high volatile C bituminous coal (Table 2-1) sized at 1/2 inch through 1 inch.

In order to study the effects of overall and bed stoichiometry, it was necessary to keep both constant during a coal run, even though the burning rate was changing significantly. This was achieved by measuring the carbon dioxide and oxygen concentration in the exhaust gases and adjusting the underfire and overfire air flows to maintain the effluent O_2 and CO_2 within 1.5% of the design point.

Excess Air Combustion

Figure 4-1 shows typical experimental results on superficial burning rate and NO_x emissions as a function of time. In this experiment, both the total and bed region stoichiometries were maintained at 1.35 (all of the air was supplied as underfire air in this case). In general, both the overall and bed region stoichiometries were controlled to within ± 0.1 of the respective average stoichiometries for the run, except in the early stages of the experiment when the burning rate was very low and high excess air conditions necessarily prevailed. In each case a control interval was defined for the run on the basis of these narrowly bounded average stoichiometries. Subsequent

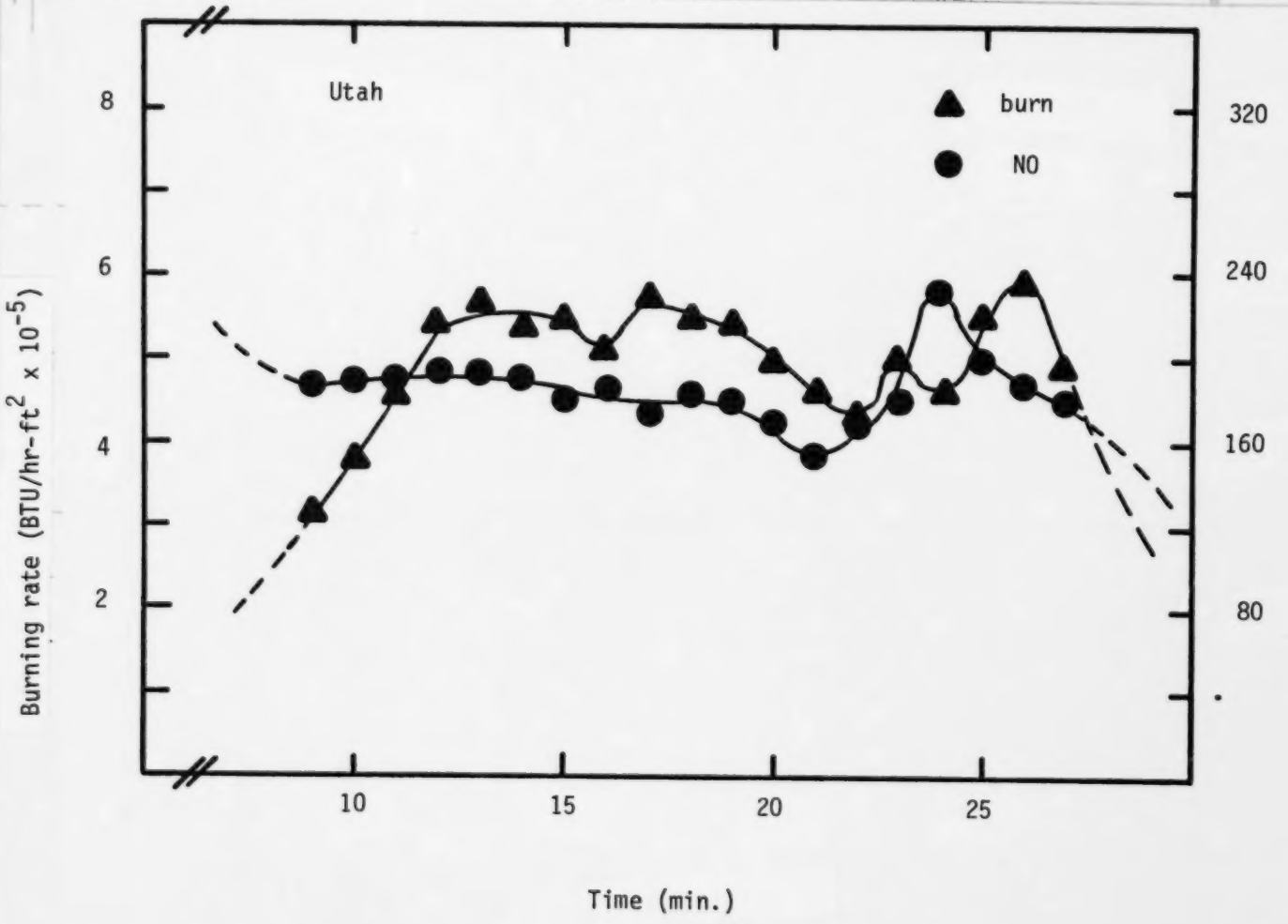

Figure 4-1. Burning rate and NO emissions for a Utah coal at SR_b = 1.35.

integrated values of NO emissions, SO_2 emissions, and burning rates were computed for the control interval only.

The burning rate data show the trends typical of the batch firing experiments. The superficial burning rate increased steadily, reached a maximum of approximately 600,000 Btu/hr ft^2 during the middle of the run, and then decreased toward the run termination. In this case, the average burning rate over the control interval (9 minutes to 27 minutes) was 598,000 Btu/hr ft^2 and this is in good agreement with commercial practice.

Figure 4-1 also shows exhaust NO concentrations converted to 0% exhaust O_2. These data indicate that under excess air conditions, NO emissions are relatively uniform throughout the control interval. This is believed to be because the nitrogenous species are evolved at approximately the same rate as the carbon species and little homogeneous or heterogeneous reduction of NO occurs at these high oxygen partial pressures.

Figure 4-2 shows the SO_2 emissions versus time for this same run. In general, the SO_2 emissions appear to follow the burning rate trends. The emissions were uniformly high (80 to 90% of theoretical) during the center portion of the test and were somewhat lower at both the run initiation and run termination where the burning rate was reduced. [Note that if the sulfur evolution perfectly paralled the carbon evolution, the SO_2 emissions (on a part per million by volume basis) would not vary with burning rate.]

Staged Combustion

In Figure 4-3a the overall and bed-region stoichiometries are given for a staged run. For this particular run the average overall stoichiometry (SR_T) was 1.39 and the bed-region stoichiometry (SR_{BED}) corresponded to a locally staged condition of 0.70.

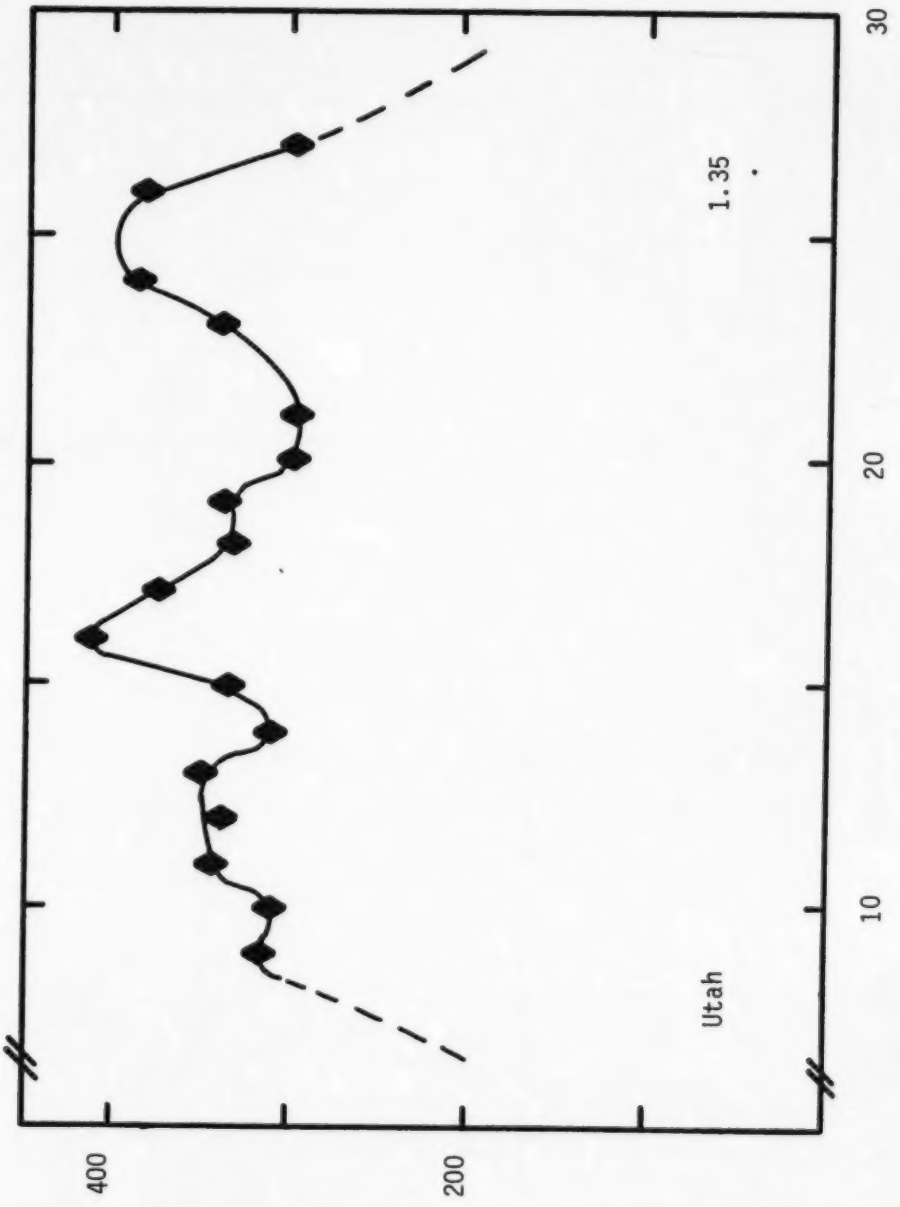

Figure 4-2. SO_2 emissions for a Utah coal at SR_b = SR_t = 1.35

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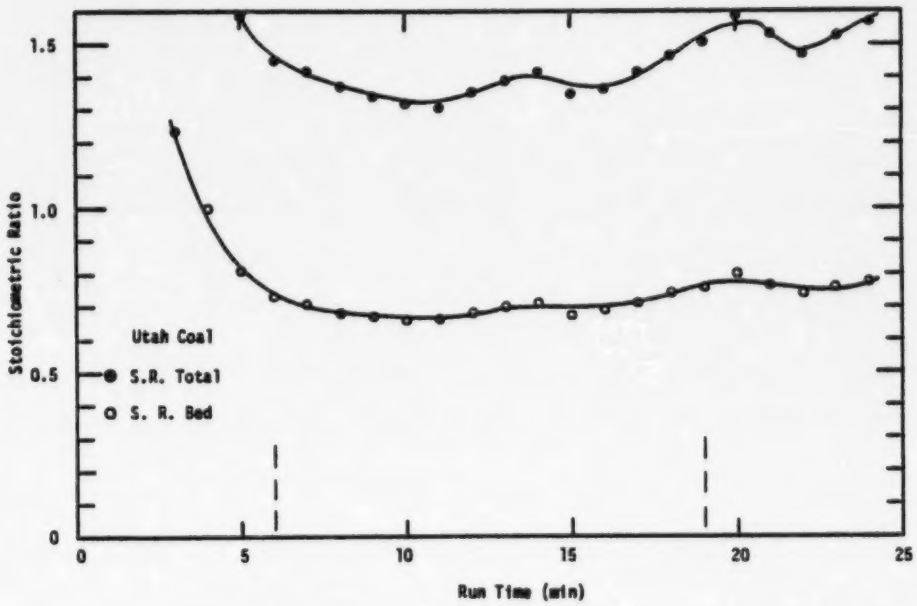

Figure 4-3a. Overall and Bed-Region Stoichiometries

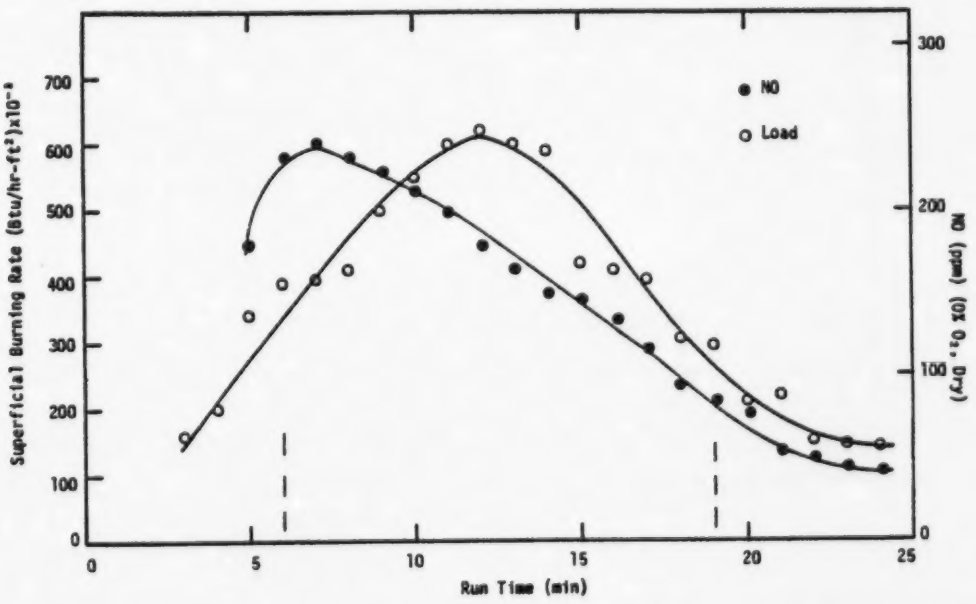

Figure 4-3b. Superficial Burning Rate and Exhaust NO Emissions

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Figure 4-3b illustrates the trends in both the superficial burning rate and NO exhaust emissions during the combustion cycle. Again, the superficial burning rate increased steadily after ignition, reached a maximum of approximately 600,000 Btu/hrft^2 at 12 minutes into the run and then decreased toward the run termination. Fuel-bed temperatures closely mirrored the burning history and suggest that the burning rate increase is attributable to an increase in local bed-temperatures resulting in a higher evolution rate of volatile matter. Optical pyrometry data indicate that under staged conditions the coal-bed burned at temperatures ranging from 2100°F to 2500°F. By contrast, under high excess-air firing of the fuel-bed (SR_{BED} > 1.20) temperatures ranged from 2700°F to 3300°F. The average coal-burning rate over the control interval (6 minutes to 19 minutes in this case) was 460,000 Btu/hrft^2 as compared with industrial chain-grate stokers observed to operate in a range from 400,000 to 500,000 Btu/hrft^2.

As shown in Figure 4-3b the NO exhaust emissions increased rapidly in the early stages of the run, passed through a maximum at 7 minutes, and then decayed toward the termination of the run. The transient behavior in NO emissions is in agreement with earlier batch coal-firing studies conducted by Haynes and Kirov.[4] The maximum in NO emissions was achieved prior to the peak in superficial burning rate (12 minutes). This "early" NO emissions peak was typical of all staged runs.

INFLUENCE OF COMBUSTION PARAMETERS

An average emission value was computed for each run by integrating the exhaust NO concentration data over the control interval (the period over which the overall and bed-region stoichiometries remained constant). The effect of control interval length upon the average NO emissions proved to be minimal

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because of the low burning rates (usually less than 200,000 Btu/hrft^2) at the beginning and end of the experiment. For all of the experiments exhaust CO emissions remained lower than 0.03 percent.

Air Distribution -- Bed-Region Stoichiometry

Figure 4-4 gives a summary of the integrated NO emissions obtained for a series of tests in which the bed-region stoichiometry was varied while the overall stoichiometry was held constant at 1.35 ± 0.03 (corresponding to an average excess oxygen of 5.6%). Utah coal was fired in these tests at an average rate of 510,000 Btu/hrft^2 ± 40,000 Btu/hrft^2. The overfire air was added in two, volumetrically equivalent portions at 15.5 inches and 44 inches above the coal bed.

Figure 4-4 shows that as the combustion was staged, with the total air supply held constant, the NO emissions decreased substantially. The data show an approximately linear dependence of NO emissions on bed-region stoichiometry. This linearity is in marked contrast to pulverized-coal combustion where NO emissions decrease sharply near a first-stage stoichiometry of 1.0 from considerably higher values at stoichiometries greater than 1.0, then level out and ultimately begin to rise again below first-stage stoichiometries of about 0.70.[18]

As expected, reducing the bed-region stoichiometry had little effect on the overall SO_2 emissions so long as the fuel was fully burned.

Air Distribution -- Excess Air Addition

Figure 4-5 summarizes the effect of overall excess oxygen on NO emissions at a constant bed-region stoichiometry. In this set of experiments the fuel-bed was fired under excess-air conditions (SR_{BED} = 1.37 ± 0.07). The overfire air was varied but distributed equally at ambient temperature between air-jet assemblies. Under these combustion conditions, the overall excess air had

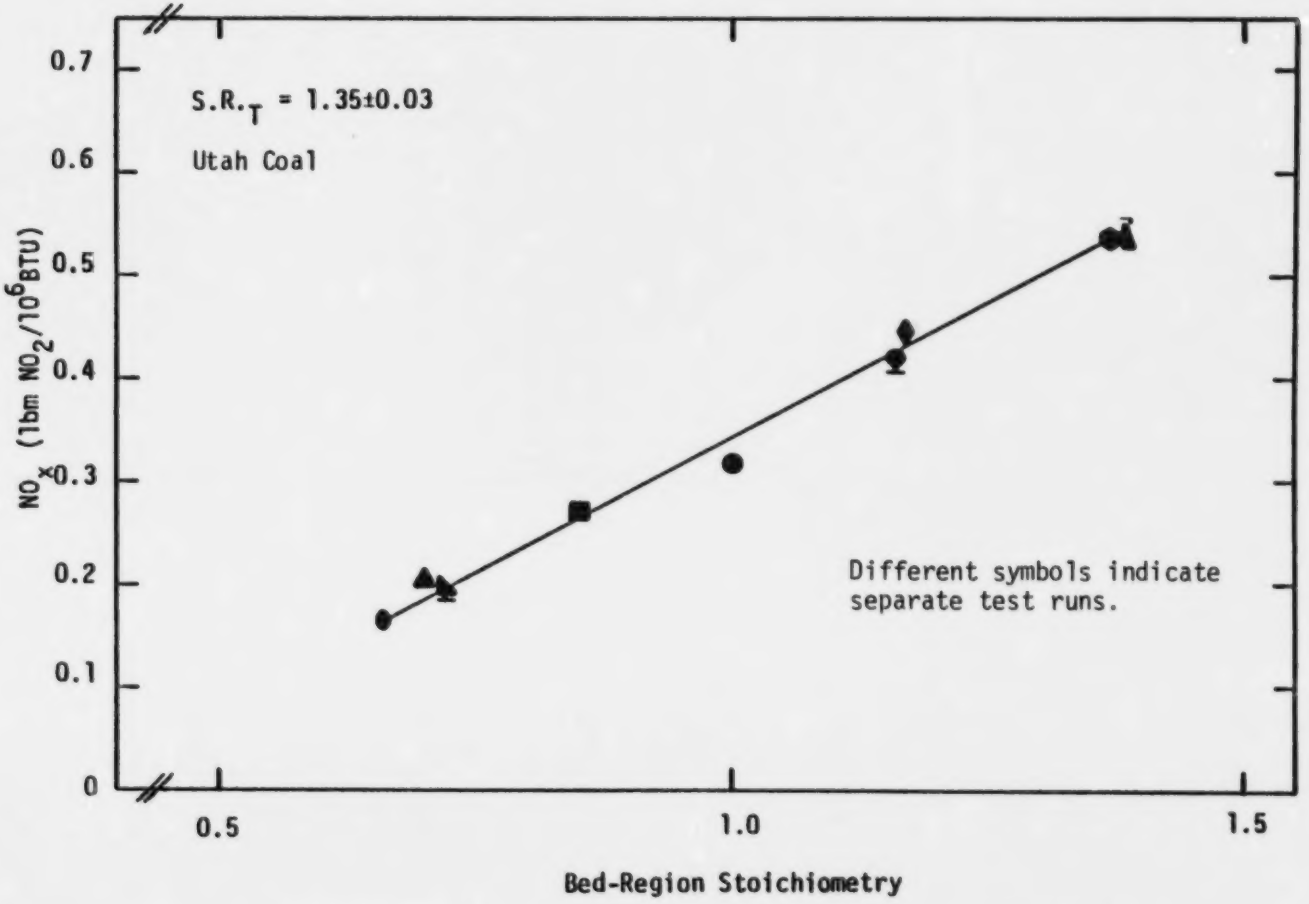

Figure 4-4. Effect of Bed-Region Stoichiometry on NO_x Emissions

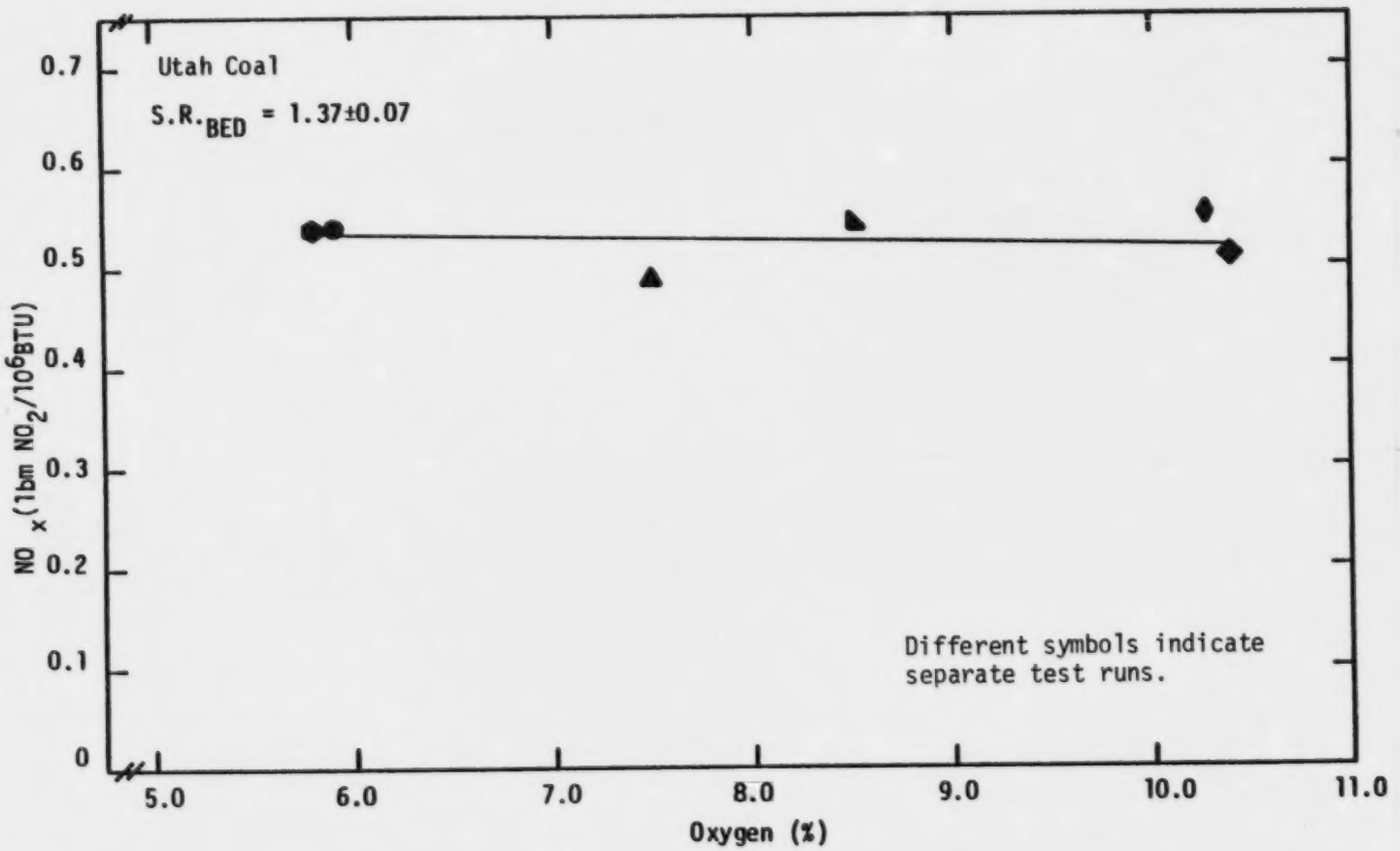

Figure 4-5. Effect of Excess Oxygen on NO_x Emissions

little effect on exhaust gas NO emissions. Excess-air firing of the fuel-bed accompanied by high exhaust oxygen concentrations is typical of industrial practice and corresponds to high NO exhaust emissions and high combustion temperatures at the fuel-bed.

Overfire-Air Height

Figure 4-6 shows the influence of stage separation upon NO exhaust emissions for two sets of bed-region stoichiometries. For the first set of experiments, the lower pair of overfire-air injectors was positioned at 9 inches above the coal bed. These air-jet assemblies were subsequently moved to 15.5 inches, 21 inches and 35 inches above the bed and the experiments were repeated. In all of the experiments the upper set of overfire-air jets remained at 44 inches above the coal-bed. The overfire air was supplied equally to the two levels of jets at ambient temperature. As previously noted, when the bed was fired under fuel-rich conditions (SR_{BED} = 0.71 ± 0.03). The NO_x emissions were significantly lower than when the bed was excess air. The overall furnace stoichiometry for these experiments was held at an average 1.40 ± 0.04. Changing the location of the lower air injectors resulted in first-stage, gas-phase residence times of 0.35 seconds, 0.80 seconds, 1.10 seconds, and 2.10 seconds, respectively.

In both sets of experiments, no effect of overfire-air height on NO emissions was observed. This absence of effect is in strong contrast to pulverized-coal combustion for which an increase in first-stage residence time under fuel-rich conditions results in reduced NO emissions, because first-stage XN species (NO, NH_3, HCN) decay toward equlibrium concentrations before entering the second-stage flame.[18]

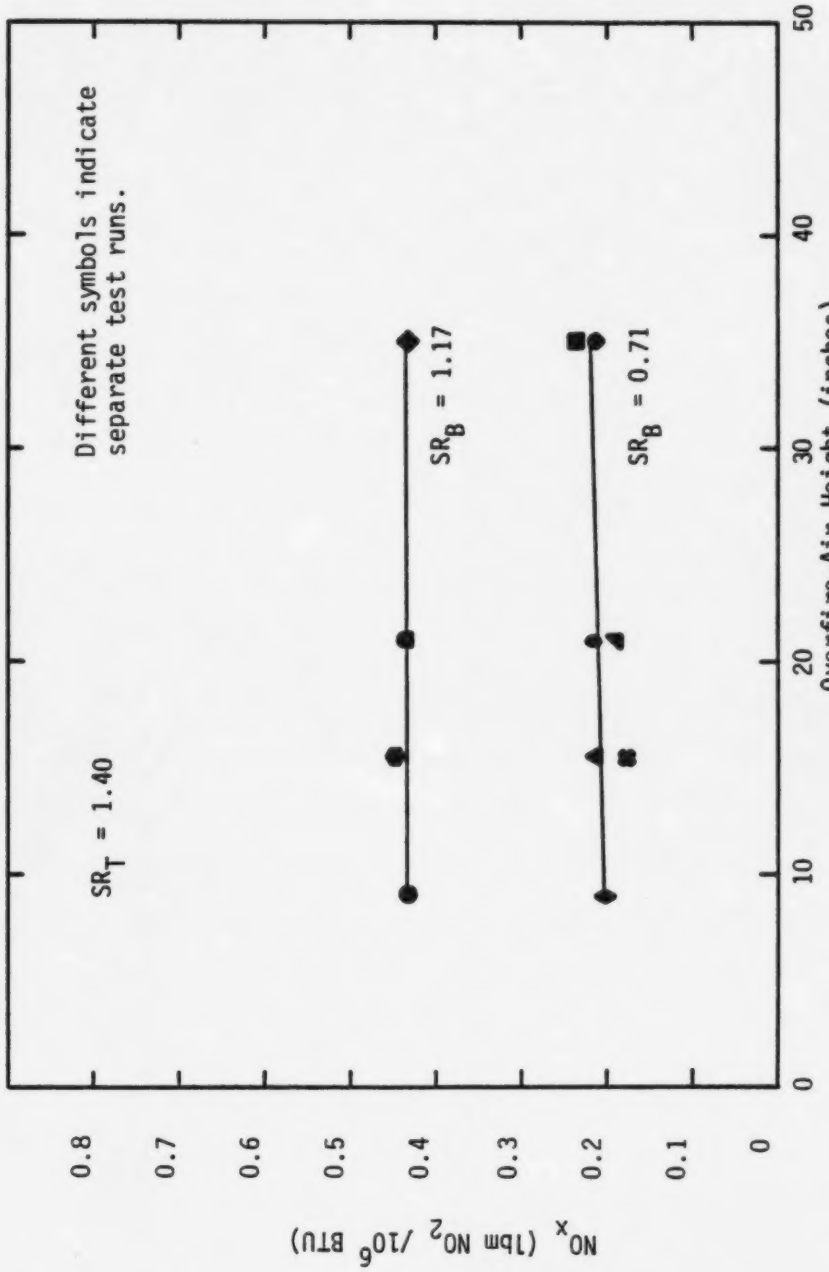

Figure 4-6. Effect of Overfire-Air Height on NO_x Emissions

Burning Rate

Johnson and coworkers[19] demonstrated the importance of first and second-stage temperature for the control of NO emissions from pulverized-coal systems; consequently, thermal effects were considered in this study.

Figure 4-7 is a summary of NO emissions data obtained from several experiments at various bed-region stoichiometries and superficial burning rates. Average NO emissions were compared over a range of constant bed-region stoichiometries, from fuel-rich to fuel-lean conditions, for widely differing fuel-bed heat-release rates. As previously noted, for all of the data-sets an increase in bed-region stoichiometry resulted in a dramatic increase in NO emissions. In all cases, within each data set NO emissions increased with increasing burning rate. While burning rate does affect NO emissions it appears to be only a second-order effect relative to local stoichiometry.

The dependence of NO emissions on bed heat-release rates suggests that as the burning rate increases, local bed temperatures increase resulting in a higher yield of volatile nitrogenous species. Pohl and Sarofim[15] have shown that the fraction of fuel-nitrogen remaining in the char depends strongly on particle temperature. As the particle temperatures are raised an increased fraction of fuel-nitrogen is evolved with other volatile species and subsequently burns homogeneously in the gas phase. A correspondingly decreased fraction of the fuel-nitrogen remains to oxidize heterogeneously with the char. Hence, increased particle (and fuel-bed) temperature may result in higher NO emissions because of higher volatile nitrogen yields.

Superficial bed heat release rate had little effect on overall SO_2 emissions; however, it did indirectly influence the sulfur evolution. Figure 4-8 shows the normalized sulfur-to-carbon evolution and oxidation profile for the Utah coal at a bed region stoichiometry of 0.7. The ordinate is the ratio

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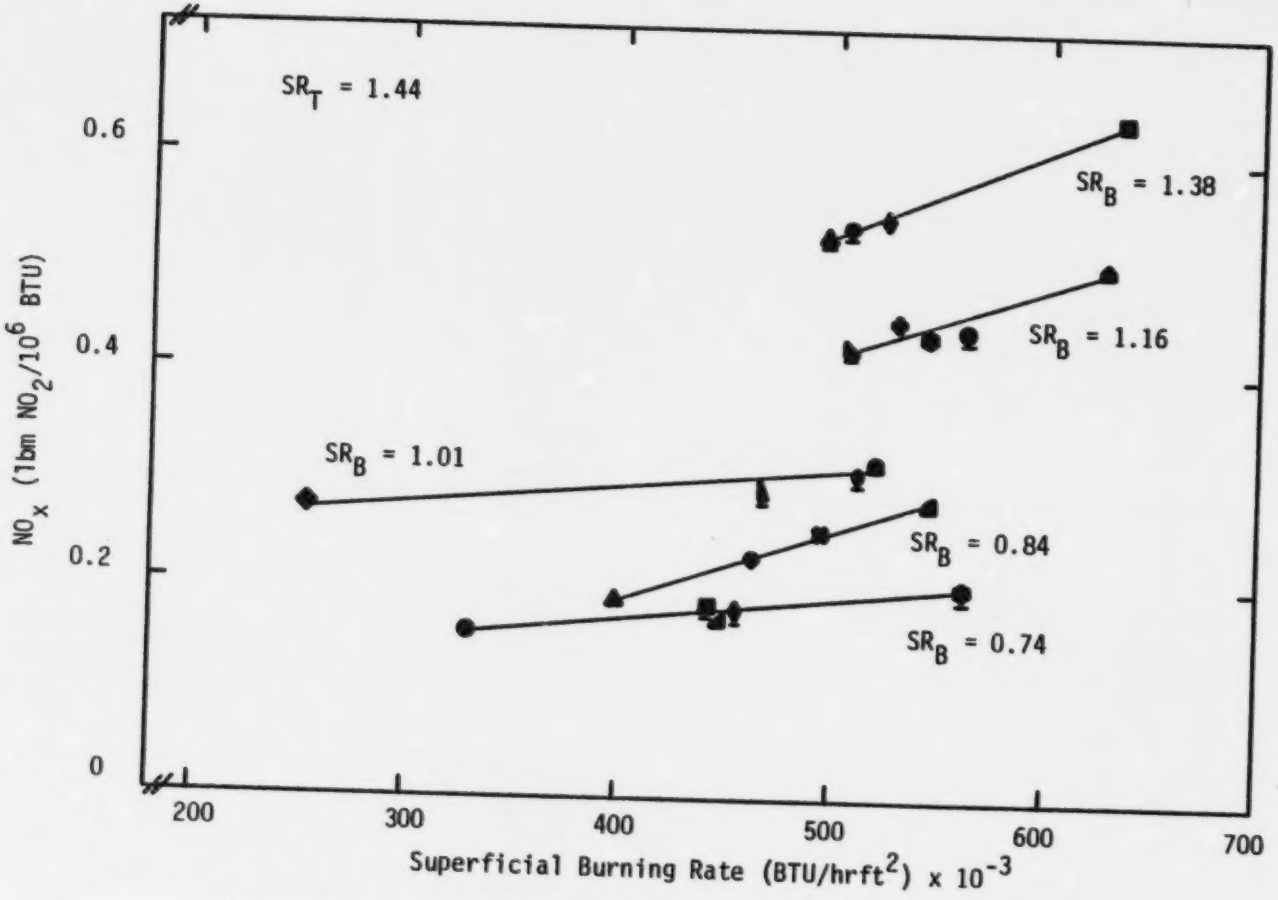

Figure 4-7. Effect of Superficial Burning Rate on NO_x Emissions

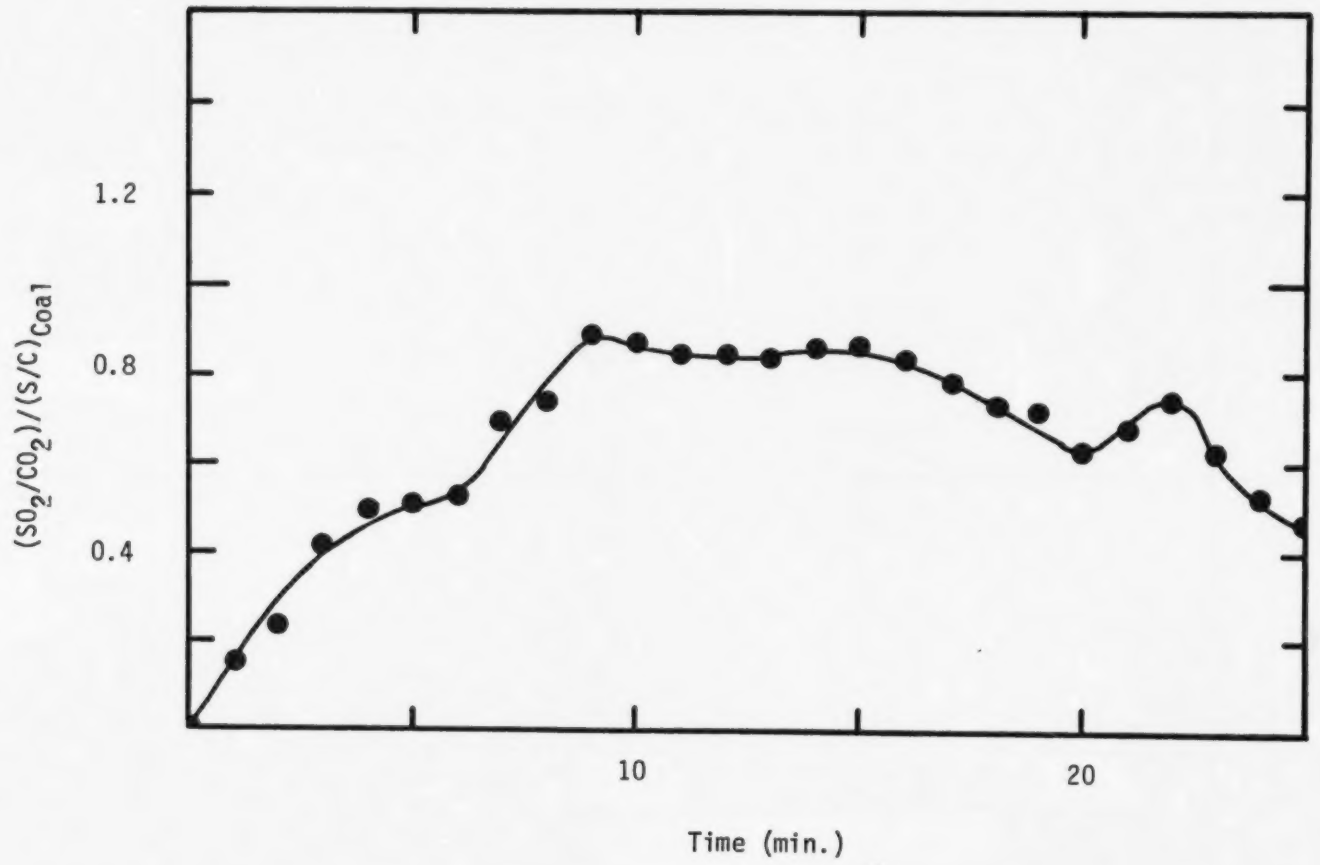

Figure 4-8. Normalized SO_2/CO_2 evolution for a Utah coal at SR_b = .7

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of SO_2 to CO_2 normalized with respect to the sulfur and carbon present in the original coal. Initially, the sulfur evolution and oxidation appears to lag the carbon burning (as indicated by a ratio significantly less than 1.0). During the main portion of the run, both sulfur and carbon oxidized at approximately the same rate. At the conclusion of the run, the sulfur oxidation again falls behind the carbon burning. As discussed earlier, these trends appear to correspond directly to the characteristic trends in bed temperature. Hence, it should be possible to influence local emission characteristics by altering the combustion profile. Figure 4-9 shows data which further support this concept. In this case, the time resolved SO_2 emissions have been plotted versus the instantaneous superficial burning rate for three separate runs with differing bed region stoichiometries. (The data have been limited to those obtained after the maximum burning rate was achieved and the bed was in full combustion.) In this regime it appears that oxygen does not strongly influence SO_2 formation; however, in general, emissions increase with increasing burning rate and hence, with increasing bed temperature. Thus, bed temperature can strongly influence the point in the run where the SO_2 formation occurs, but ultimately the sulfur will be evolved and oxidized producing the same overall emissions.

COAL-LIMESTONE PELLETS

Limited testing was conducted with coal-limestone pellets obtained from Battelle Memorial Institute to determine the influence of combustion parameters and the effectiveness of sulfur capture by adding limestone. Table 4-1 gives the ultimate composition and ash analysis of the pellets. The parent coal was a mid-western bituminous fuel with approximately 3% sulfur. Piqua limestone was sulfur molar ratio 3.5. Because of the limestone, the coal-limestone pellets were significantly lower in calorific content (4,720

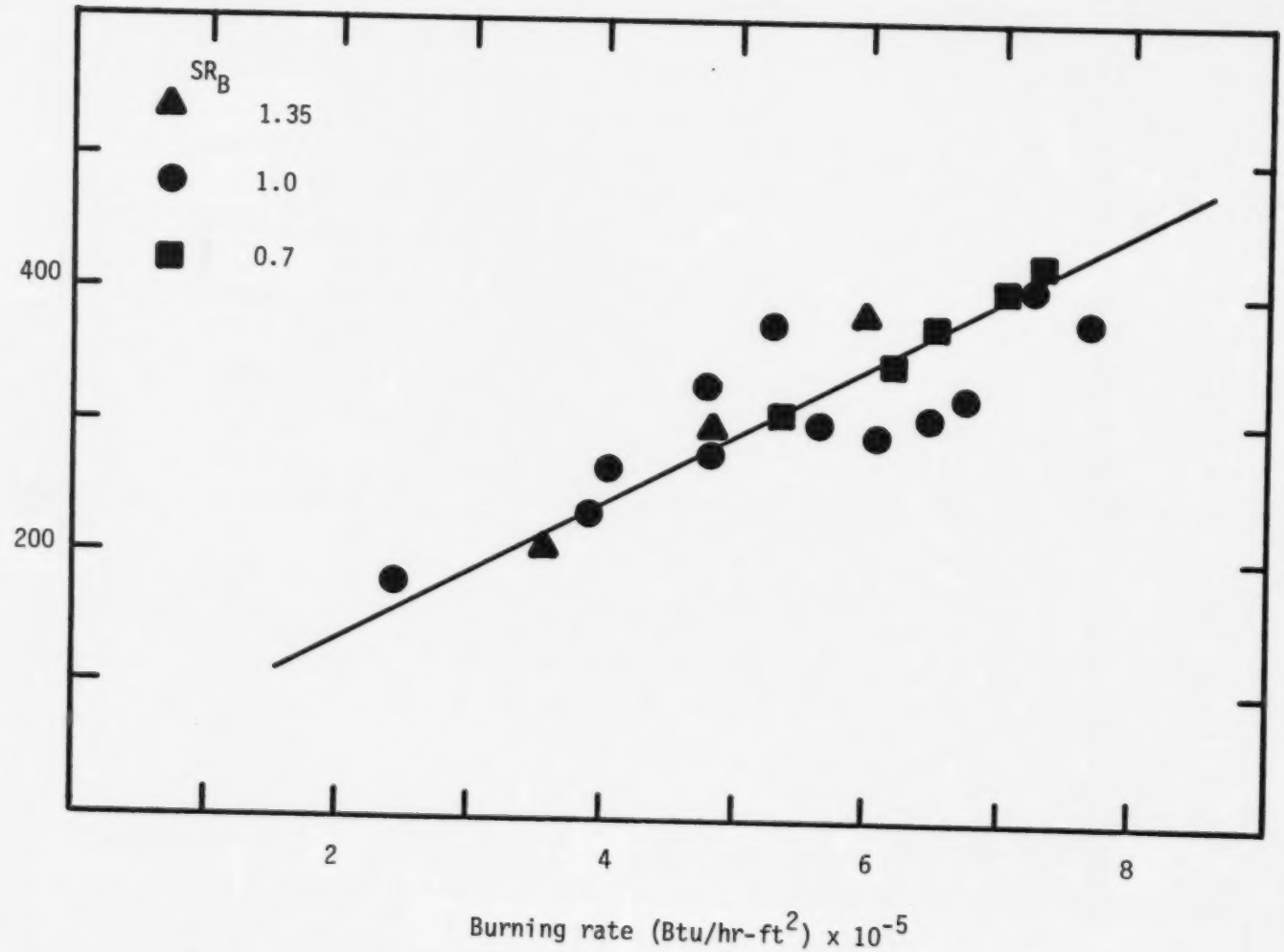

Figure 4-9. Post maximum burning rate correlation for various bed stoichiometries

Btu/lb) than the previously used Utah coal (11,658 Btu/lb). The experimental test procedures were identical to those used previously in firing the more conventional coals. Again the fines were removed prior to firing in the fixed-bed furnace.

Figure 4-10 shows a typical firing rate versus time response curve for a bed region stoichiometry of 1.0 and an overall stoichiometry of 1.4. The data shown in Figure 4-10 are typical and indicate that in general the combustion tests were of shorter duration and the maximum heat release was less than in the case of the Utah and other bituminous coals.

The pellets themselves appeared to burn similar to the bituminous coals. They were approximately 1 inch long and 1/2 inch in diameter. The limestone pellets maintained their integrity almost throughout the entire run; only near the end did they crumble into indistinctness. The ash was easily handled at all test conditions; operation with bed region stoichiometries less than 1.0 did not result in the formation of large clinkers.

Table 4-1

Ultimate Analysis of Battelle Pellets

Pellet Composition	
Coal Moisture	12.60%
Carbon	29.70%
Hydrogen	1.54%
Nitrogen	0.48%
Sulfur	1.96%
Oxygen (by difference)	8.64%
Coal Ash	45.60%
Caloric Heating Value (Btu/lb)	4720

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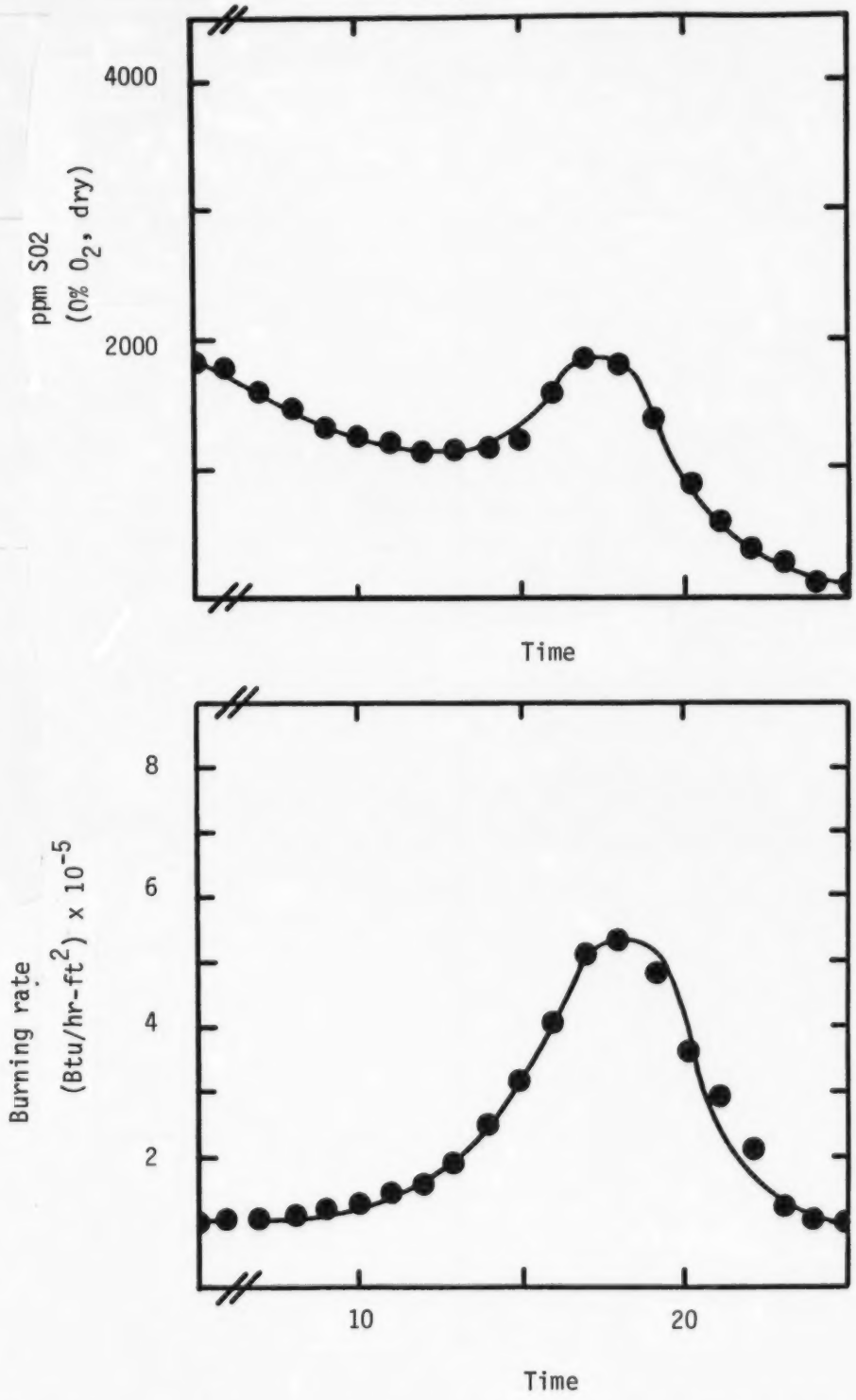

Figure 4-10. SO_2 emissions and burning rate for Battelle pellets

Figure 4-10 also shows the SO_2 emissions for this typical run, and indicates that significant sulfur capture was observed throughout the test. As in the previous coal runs, the SO_2 emissions followed the heat release rate, but the results were damped as a result of the sulfur capture. The data shown in Figure 4-10 indicate that the poorest capture occurs early in the run (high oxygen concentrations) and at the peak heat release rate. The latter effect is believed to be directly attributable to the increased local particle temperatures at this point in the run.

Figure 4-11 summarizes all of the results obtained with the Battelle pellets by presenting the integrated SO_2 emissions as a function of bed region stoichiometry. In general, the data were found to be very reproducible and indicate that total sulfur emissions decrease slightly as the stoichiometric ratio in the bed is reduced. Sulfur retention under fuel lean (excess air) combustion conditions was quite good as indicated by Figure 4-11 (almost 70% at a Ca/S ratio of 3.5). However, under fuel rich conditions the capture increased to over 80%. No attempt was made to provide additional cooling to the bed. These tests were conducted under conditions identical to those used for the evaluation of the five bituminous coals. However, it should be noted that the pellets have an inherently lower burning rate due to the added limestone and hence, the bed temperatures were somewhat reduced naturally.

CONCLUSIONS

NO Formation and Control

The experimental studies conducted with the transient mode, fixed-bed firing system have shown that NO formation is not uniform throughout the burning time of the fuel-bed. The formation maximizes early in the process, prior to the peak combustion rate and then gradually decreases toward the termination. Staged-air addition is a potentially effective means of NO

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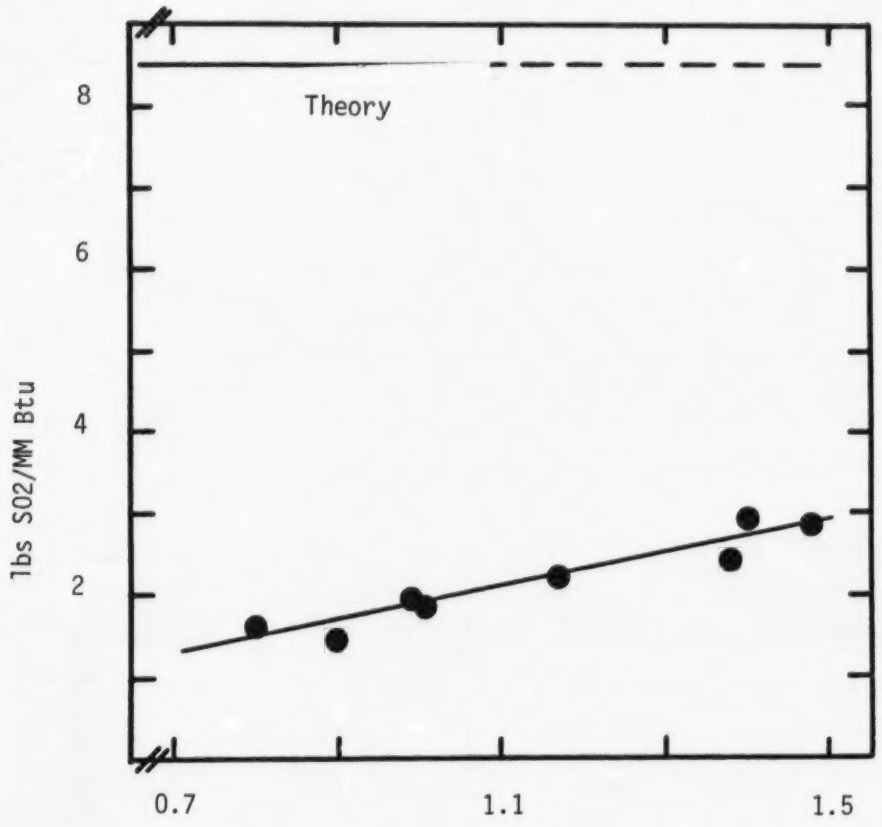

Figure 4-11. Integrated SO_2 emissions for batch mode burning of Battelle pellets.

control. Exhaust concentrations decrease approximately linearly with decreasing bed stoichiometry. Overall stoichiometry appears to have little effect on NO emissions under staged conditions. Overfire air height (first-stage-residence time) is not of first-order importance with respect to NO emissions. An increase in superficial heat-release rate increases the NO emissions under both staged and excess-air conditions.

SO_2 Formation and Control

The results of this experimental research have suggested that sulfur evolution and oxidation are not uniform throughout the burning time of the bed. In general, sulfur oxidation appears to lag the carbon burning, during the initial and final stages of the bed combustion. Local emission levels in excess of the theoretical concentration are possible due to the evolution of inorganic sulfur in peak temperature regimes. SO_2 emissions correlate well with the burning rate (and hence bed temperature) during the middle and later stages of burnout. Overall and bed region stoichiometries appear to have little effect on total SO_2 emissions so long as the fuel is fully burned. Utilization of coal-limestone pellets can result in significantly reduced SO_2 emissions. The sorbent effectiveness appears to increase when the bed region stoichiometry is reduced below 1.0.

CHAPTER 5

MODEL SPREADER-STOKER STUDIES

COMPARISON WITH FIELD DATA

Experiments were conducted in the model spreader stoker furnace (MSF) to investigate the effect of both the bed phase and suspension phase combustion, and to assess the overall influence of the stoichiometry distribution and overfire air heights on NO formation and control. Initial validation tests were conducted on the MSF to compare NO formation in the pilot-scale facility with emission data from full scale units. Typical NO emissions data for full-scale boilers are shown in Figure 5-1 by the solid lines. Each line refers to a specific furnace from the work of Langsjoen, et al.[3] While the exact bed region stoichiometries are unknown in these field tests, they can be approximated by the use of the general operating characteristics.

Nitric oxide emissions data from the model spreader stoker unit, operated under conditions typical of full scale practice, are also shown in Figure 5-1 (solid points). These tests were conducted at a firing rate of 470,000 Btu/hr. Overfire air represented 18 percent of the total air supplied to the furnace. The overfire air was introduced at 20 and 48 inches above the grate; the ratio of the air introduced at 48 inches to that at 20 inches was maintained constant at 1.15. The quantitative agreement between the pilot scale model spreader stoker emissions and the field testing data suggest that the pilot furnace appropriately simulates the combustion conditions which are of primary importance to NO formation.

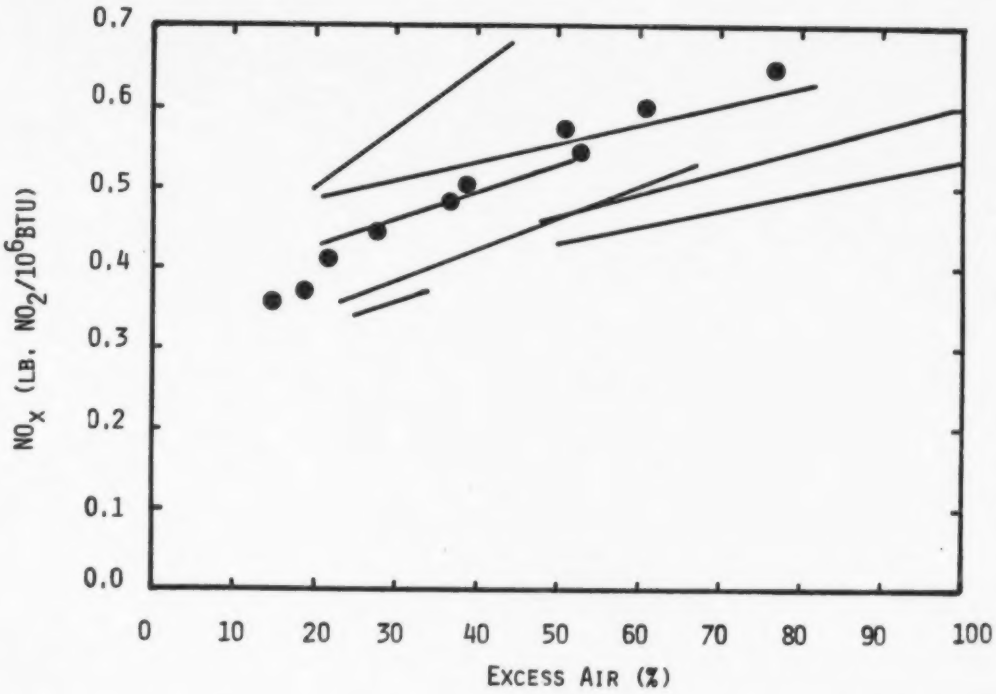

Figure 5.1. Field Data Comparison

TOTAL STOICHIOMETRY

In further tests, the effects of bed zone stoichiometry and overall stoichiometry were studied. Figure 5-2 shows NO emissions for a series of experiments in which the bed zone stoichiometry was held constant at an excess air level of 18%; the coal feed rate was 29 lbs/hr., which corresponds to approximately 350,000 Btu/hrft^2. The experiment was performed at two separate height levels of overfire air, 20 and 48 inches. As the amount of overfire air was varied to produce different overall stoichiometries, the level of NO emissions remained essentially constant. These results indicate that exhaust NO emissions are relatively insensitive to overall stoichiometry as long as the bed region stoichiometry is held constant. These data are consistent with similar pulverized coal results and indicate a weak dependence on oxygen partial pressure under excess air conditions.

BED-REGION STOICHIOMETRY

The effect of bed stoichiometry was studied at 470,000 Btu/hr under conditions such that the overall stoichiometry was held constant at 1.45. As the air/fuel ratio in the bed region was reduced by putting a larger fraction of the air through overfire air jets, the NO emissions decreased significantly as shown in Figure 5-3. When the bed was operated with substantial excess air (SR_{BED} > 1.3) the visual flame zone was confined to approximately 6 inches above the bed. However, as the bed stoichiometry was decreased, the height of the volatile combustion zone increased significantly and by SR_{BED} = 0.65 was on the order of 3 feet. Prior to these tests there had been serious concern about the operability of a continuous feed system at sub-stoichiometric conditions with coal firing; however, no combustion problems were encountered. The extent of the reduction in NO emissions when the bed was operated under sub-stoichiometric conditions was also a function of the height of the

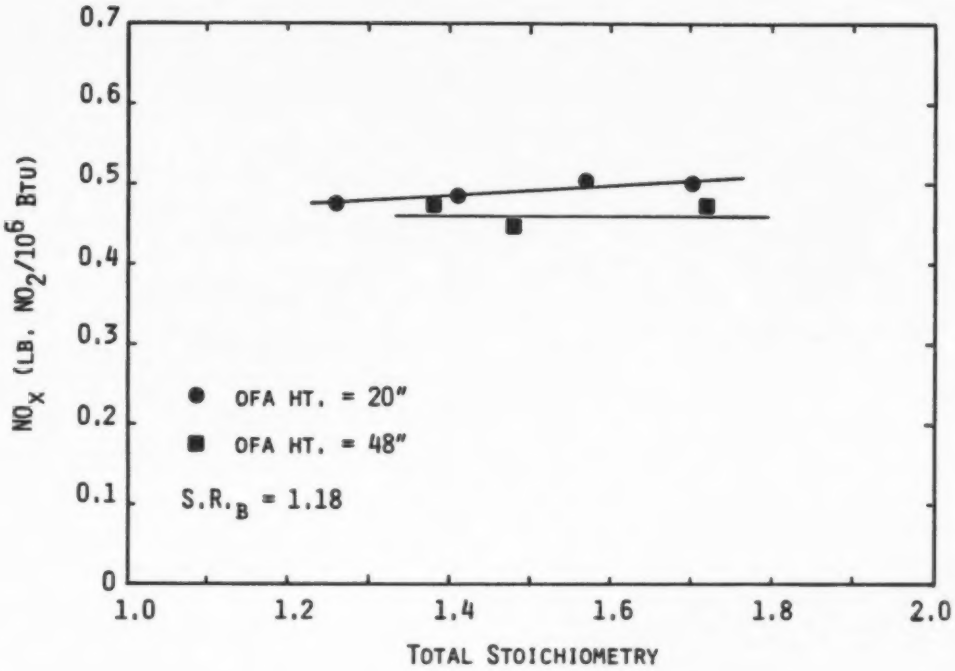

Figure 5.2. Effect of Overfire Air

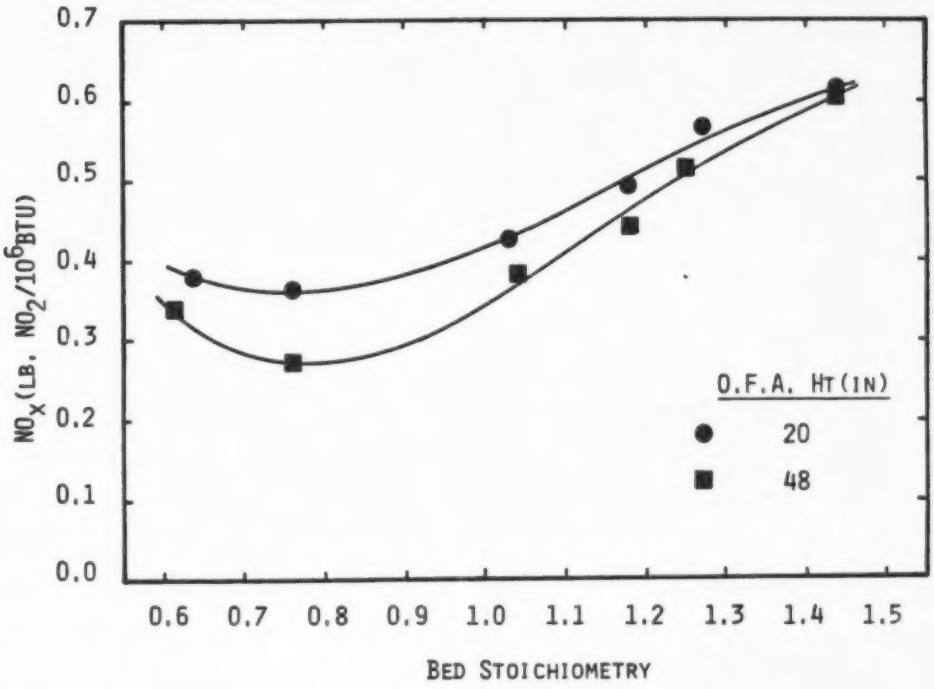

Figure 5-3. Effect of Bed Stoichiometry

overfire air jets above the bed. This difference is an indication of the effect that suspension zone oxygen levels exert on exhaust NO emissions. When the overfire air was injected at 20 inches above the grate, (i.e. below the level of fuel injection at the spreader), then much of the suspension zone combustion occurred under an excess air condition. Lower levels of NO were produced when the overfire air was injected at 48 inches above the grate, thus enclosing the suspension zone in a fuel-rich, oxygen deficient environment.

OVERFIRE AIR HEIGHT

The strong influence of overfire air height on NO emissions is shown in Figure 5-4. In these experiments, the height of overfire air injection was varied from 7 to 72 inches above the grate. Injecting the overfire air above the spreader during staged combustion enhances the probability that fuel N evolved from the small particles burning in the suspension phase will react to form N_2. In addition, increasing the overfire air height increases the rich zone residence time and allows gas phase nitrogen species leaving the fuel bed to decay further toward their low equilibrium levels.

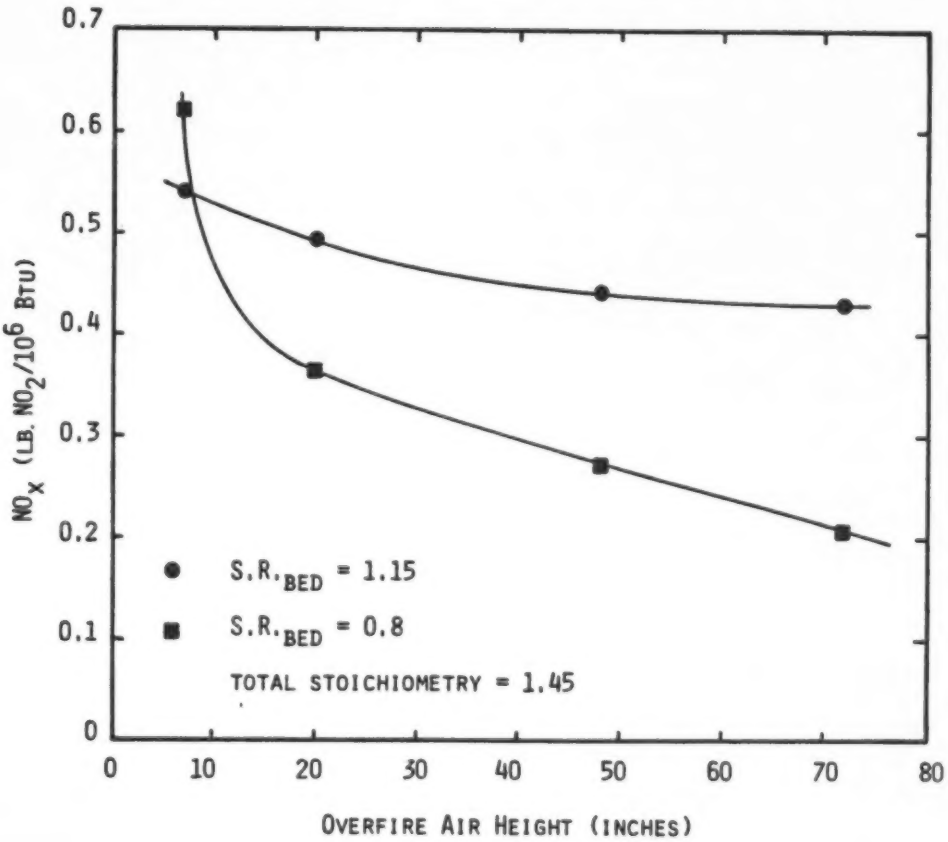

Figure 5-4. Effect of Overfire Air Height

CHAPTER 6

DISCUSSION

The results of this study indicate that NO emissions from spreader stoker fired coal furnaces are the result of relatively high conversions (25-45%) of the fuel nitrogen evolved from particles less than 0.11 inches in the suspension phase and low conversions (5 to 15%) of fuel nitrogen from the bed combustion. Mass burning stokers (e.g. traveling grate units) almost certainly yield lower NO_x emissions because of the absence of significant overthrow combustion. In the suspension phase, the nitrogen is evolved at approximately the same rate as the carbon is oxidized and the percentage conversion is relatively insensitive to reaction zone temperature. Particle size also has little effect on the percentage conversion, but it dramatically influences the amount of fuel nitrogen evolved.

Local oxygen availability is the primary control parameter for both phases of the combustion. Substantial NO_x emission reductions were achieved when the fuel bed was operated under sub-stoichiometric conditions (fuel rich). NO_x emissions decreased further when the stoichiometry of the suspension zone was reduced below 1.0 by moving the overfire air injection location above the spreader. Under these conditions a minimum overall fuel nitrogen conversion of approximately 6 percent was achieved suggesting that under staged combustion conditions, a positive synergism between the suspension phase and the bed combustion can be achieved. The suspension zone almost certainly provides both sites for heterogeneous reduction of NO formed in the

bed and the radical concentrations necessary to accelerate the decay of TFN formed in the bed. In addition, the low oxygen partial pressure decreases the conversion of nitrogen evolved in the suspension zone.

Thus, two types of NO_x control technology appear to be viable. First, the coal feed could be screened to remove the fines (particles less than 0.1 inches). This would essentially eliminate the high conversion of nitrogen evolved in the suspension zone and would result in an overall emissions reduction of approximately 10 to 40 percent depending on the amount of fines normally present in the raw coal. Alternatively, the primary overfire-air injection port could be moved to above the spreader and the suspension zone and bed region could both be operated sub-stoichiometric. This could result in an emission reduction as large as 50 percent; however, before this concept can be applied commercially, the influence on other important combustion parameters (e.g. bed thickness, grate temperature, and ash characteristics) must be evaluated.

Unfortunately, control of SO_2 formation in either a spreader or mass-burning stoker unit appears to be difficult. The rate of sulfur evolution and oxidation in both the suspension and fixed-bed phases can be influenced by the local temperature; however, if the coal is completely burned, the sulfur is ultimately oxidized to SO_2. Utilization of coal-limestone pellets can result in significantly reduced SO_2 emissions and the effectiveness of the sorbent appears to increase when the bed-region stoichiometry is reduced below 1.0.

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